



## Review Article

# Utilization of Tannins with Various Polymers for Green-Based Active Packaging: A Review

Fidela Devina Agrippina<sup>1,\*</sup>, Maya Ismayati<sup>2</sup>, Sri Hidayati<sup>1</sup>, Bima Putra Pratama<sup>3</sup>

<sup>1</sup> Department of Agro-Industrial Technology, Faculty of Agriculture, University of Lampung, Bandar Lampung, Indonesia

<sup>2</sup> Research Center for Biomass and Bioproducts, National Research and Innovation Agency (BRIN), Bogor, Indonesia

<sup>3</sup> Research Center for Agroindustry, National Research and Innovation Agency (BRIN), Bogor, Indonesia

\* Corresponding Author. E-mail address: [fidela.devina.agrippina@gmail.com](mailto:fidela.devina.agrippina@gmail.com)

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## ABSTRACT

Tannin is a major polyphenolic secondary metabolite widely distributed in the forestry sector and can be added to polymer matrices to manufacture bioactive films for food packaging. Tannins have functional properties as antioxidants, antibacterial, and anti-inflammatories. Tannins are classified into two types, namely condensed tannins and hydrolyzable tannins. Tannins have been used primarily in food, wood, leather, pharmaceutical, and other industries. In the food industry, tannins are used to develop food packaging, preservation, and the function of the food industry. Tannin molecules have the advantage of combining with polymers or polymer surfaces, for example, synthetic polymers, biopolymers, and micro- and nano-sized fibers, which will form new products whose physical and chemical properties increase in functional properties because of the presence of hydrophilic and nucleophilic groups. Combining tannins with a polymer can be done by one-step reactive extrusion, layer by layer, dynamic vulcanization, acetylation, and in situ extraction methods. This paper describes general information about tannins, followed by applications using tannins, tannin-based hybrid materials, and methods of combining tannins with a polymer for food packaging purposes.

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## 1. Introduction

Over 1.3 billion tons of human food products are thrown away yearly (Gustavsson et al. 2011). This tendency is because of poor harvesting, transportation, and storage procedures (Morris et al. 2017). This massive volume of food waste places a significant financial burden on the food business. Therefore, developing suitable packaging for perishable commodities is crucial to allow more extended transport and storage times, thus increasing shelf life (Lloyd et al. 2018). However, designing and manufacturing suitable intelligent packaging technologies is challenging from an industry perspective for various reasons, such as antimicrobial and active packaging.

Active packaging has become increasingly popular due to its ability to prolong shelf life and keep the quality of fresh fruit and vegetables. The total market for developed active packaging was estimated at around USD 1.3 billion in 2020. It is projected to expand at a compounded annual growth rate (CAGR) of 8.5% and reach USD 2.5 billion by 2027 (Peng et al. 2019). According to Fuertes et al. (2016), the market for this kind of packaging is predicted to increase at a CAGR of 7.4% in the US and reach USD 3.6 billion over the next ten years. Japan is the second-largest

market, valued at USD 2.36 billion; Australia at USD 1.69 billion; and Great Britain at USD 1.27 billion. Moreover, Germany provided the equivalent of USD 1.4 billion.

Biodegradable active packaging made from bio-based components is becoming increasingly important to handle the environmental impacts of food and packaging waste. These materials are more suitable for applications involving food contact, with fewer issues relating to harmful chemical migration (Geueke et al. 2018). In addition, the current customer demands using natural products that do not contain chemical additives for food preservation. Due to this, there is a growing need for natural active ingredients that can enhance food shelf life (Li et al. 2024). Since the active ingredients can maintain their initial mechanical and barrier properties, further development of these materials is increasingly important (Perera et al. 2023). According to Realini and Marcos (2014), the use of biodegradable packaging materials, such as carrier polymers and active compounds obtained from natural sources like essential oils (Mohamed et al. 2020), polyphenols (Ali et al. 2019), flavonoids (Przybyłek and Karpiński 2019), tannins (Zhai et al. 2018), coumarin (Benbettaieb et al. 2016), and quinone (Bouarab Chibane et al. 2019), will continue to increase.

In this context, phenolic compounds produced from plant extracts, especially tannins, are being studied as prospective components for incorporation into polymer matrices to make food packaging bioactive films (Cano et al. 2020). Tannin is a major polyphenolic secondary metabolite widely distributed in the forestry sector and is found mainly in bark, stems, seeds, roots, buds, and leaves (Giovando et al. 2019; Tomak and Gonultas 2018). The forestry sectors with higher condensed tannins are mangrove, quebracho, hemlock, and wattle. On the other hand, chestnuts and myrobalan exhibit higher levels of hydrolyzable tannins (Das et al. 2020). They are known for their biophysicochemical properties, including ultraviolet (UV), antioxidant, antimicrobial, and antifungal properties (Pizzi 2019a; Pratama et al. 2022a). Antioxidants can prevent oxidation reactions by binding to free radicals, slowing down the process of decomposition and rancidity. Based on their chemical characteristics, “tannin” refers to two groups of phenolic compounds: condensed tannins and hydrolyzable tannins (Peng et al. 2013). Combining tannins and biodegradable polymers is one of the best methods for producing novel materials with the appropriate properties.

In addition, they have antioxidant, antibacterial, antifungal, and antiviral properties (Zhai et al. 2018). Packaging films incorporating plant extract into polymers often change physicochemical, mechanical, and barrier properties compared to films made of individual components. These films have been used for various polymer applications (Mir et al. 2018). In recent years, it has been demonstrated that tannins can increase the shelf life of polymeric materials by reducing thermal oxidative action and UV degradation (Bridson et al. 2015; Shnawa 2017). Tea extracts (Chenwei et al. 2018; Peng et al. 2013; Wen et al. 2020), gallic acid and quercetin (Luzi et al. 2019), tannic acid (Dai et al. 2018) and wood or bark tannins (da Cruz et al. 2020; Zhai et al. 2018) have been reported to associate with PVA, exhibit antioxidant activity, and an antimicrobial film made by releasing antioxidants after application. Condensed tannins are more appealing for the production of active packaging materials than the other tannins described above due to their high availability (more than 90% of commercial tannin production globally) and relatively low price (especially those derived from bark) (Arbenz and Avérous 2015). Condensed tannins have mainly been used in the adhesive industry as a source of phenols over the past century, and they are used more in traditional ways, like tanning leather (Pizzi 2019a). However, condensed tannins have not been widely utilized in producing active packaging materials. Therefore, this literature

review is to explain more about tannin sources, tannin applications, and tannin preparation methods combined with polymer as active food packaging. Conclusively, emerging applications are displayed, and future perspectives are described.

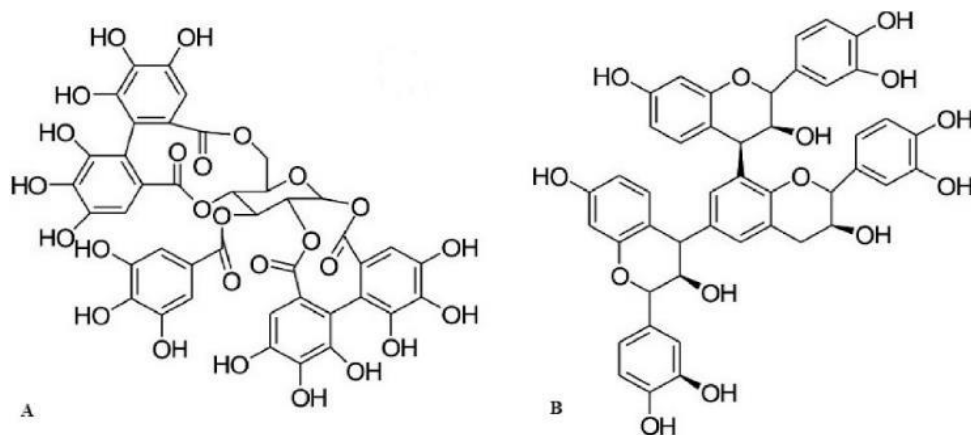
## 2. Tannin

### 2.1. Definition of Tannin

Tannins are secondary metabolites present in plants that plants synthesize. Its availability in the plant is quite diverse in the number of percent but spread in parts in almost all plants (Pizzi 2019b). Tannins have a molecular weight of 500–3000 with a large number of phenolic hydroxyl groups, which enable them to cross-link many different molecules, including polysaccharides, amino acids, fatty acids, and nucleic acids, effectively (Schröpfer and Meyer 2016). The presence of phenolic hydroxyl groups in tannins can cause tannins to have functional properties, such as antioxidants, antibacterial, and anti-inflammatory (Houston et al. 2017). It is explained in the reference that tannin's antioxidant activity, on the chemical side, is an electron-donating compound that inhibits the damage caused by free radical compounds. Antioxidants prevent the chain reaction leading to the free radical generation and stabilize free radicals by making up for the free radicals' electrons (Hayat et al. 2020; Pratama et al. 2022b). Tannins include many phenol rings, specifically hydroxyl groups linked to aromatic rings, making them easily oxidized by contributing hydrogen atoms to free radicals (Benzidia et al. 2019).

### 2.2. Types of Tannin

Tannins have been classified into hydrolyzable and condensed tannins (Fig. 1). Hydrolyzable tannins often contain carbohydrates (commonly d-glucose) at the core of their structure. Meanwhile, phenolic groups, including gallic acid in gallotannin and ellagic acid in ellagitannins, partially or entirely esterify the hydroxyl groups of carbohydrates in tannins. Under the aid of weak acids or bases, the hydrolyzable tannins can be converted into sugars and phenolic acids. The second type, condensed tannins, or proanthocyanidins, are polymers of 2 to 50 or more flavonoid units linked by carbon-carbon bonds that cannot be separated using hydrolysis processes. In terms of characteristics, hydrolyzed tannins and some condensed tannins are water-soluble, but most condensed tannins are not (Okuda and Ito 2011).



**Fig. 1.** Structure of tannins: A. Hydrolyzable tannins and B. Condensed tannins (Raja et al. 2014).

### 2.3. Sources of Tannin

Tannins can be found in several sources, such as tree bark, seeds, leaves, fruit, roots, and rhizomes. On tree bark, tannins are found in the bark of acacia mimosa, cinnamon, cinchona, wild cherry, willow, and several other species, such as oak and hamamelis. In addition, in seeds such as cocoa, areca, guarana, and kola. In leaves, tannins are found in hamamelis and green tea leaves. Meanwhile, tannins are found in the rhizomes of krameria roots (rhatany) and the roots of fern plants. Furthermore, it can be found in fruits such as *Diospyros kaki* and *Quercus infectoria*. Tannins are relatively evenly distributed in certain parts of almost all types of plants. Therefore, the potential and availability that are easily obtained make tannins a potential source of natural ingredients to be utilized by humans (Bele et al. 2010). The sources of condensed and hydrolyzable tannins are summarized in **Table 1**.

### 2.4. Applications of Tannin

Crude oil-based raw materials have been exploited in many fields and industries. This causes very high demand and soaring price increases, and the availability of raw materials is increasingly running low. In addition, using petroleum as a polymer-based material raises many challenges and drawbacks because the processing process produces large amounts of CO<sub>2</sub> emissions, which impact the environment, other living things, and human health (Huang et al. 2022). Therefore, a breakthrough to produce similar products from renewable raw materials, natural biopolymers, is needed. Several biopolymers are being developed and can potentially replace hydrocarbon biopolymers, including polylactic acid, chitosan, lignin, and tannins derived from plants and animals. Tannins are superior biopolymers among those mentioned because of their nature, structure, and abundance, making tannins usable with excellent economic value (Gómez-Plaza et al. 2016). Tannins can be employed in various industries, including food, pharmaceutical, wood composites, leather, and others, which are further explained as follows:

#### 2.4.1. Food industry

Tannins belong to the polyphenol group found in many natural plants or plant materials. The advantage resulting from its structure, which has a hydroxyl group, is that tannins have a positive effect as an antioxidant, antibacterial, and antifungal, which a food ingredient requires to produce to reach the consumer. Therefore, tannin compounds can be used in the food industry, which can be modified into food packaging materials, and their active compounds can be used as food preservatives.

##### 2.4.1.1. Food packaging

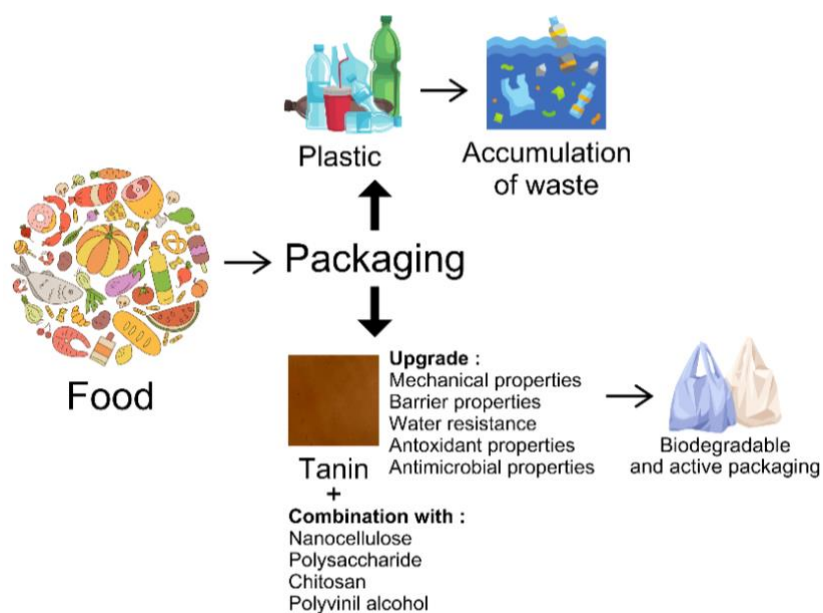
So far, the packaging of food ingredients uses plastic, polyethylene, and the like; because they are considered light, the manufacturing process is relatively easy, and their availability is also quite abundant. However, the use of plastic provides a very high level of environmental pollution. These materials are also tough to degrade by nature, thus creating global pollution (Thushari and Senevirathna 2020). Therefore, innovation is needed in packaging for food products easily degraded by nature and can be as good as the previous packaging. Tannins can be used and embedded in a biopolymer material so that their physical abilities can approach plastic (Ferri et al. 2023). They are added to the acting ability of tannins and are then referred to as bioactive

packaging that is easily degraded (Grigsby et al. 2015). One example is the packaging of nanocellulose-tannin base material (Fig. 2) (Missio et al. 2018).

**Table 1.** The sources of condensed and hydrolyzable tannins

Type	Parts of Plant	Source	References
Condensed tannins	Barks	Cinnamon	(Mateos-Martín et al. 2012)
		Mimosa ( <i>Acacia mearnsii</i> )	(Panzella and Napolitano 2022)
		Pine ( <i>Pinus pinaster</i> )	(Panzella and Napolitano 2022)
		Quebracho ( <i>Schinopsis balanse</i> )	(Panzella and Napolitano 2022)
		Mangrove ( <i>Rhizophora mucronata</i> )	(Pancapalaga et al. 2023)
		Mangrove ( <i>Maclura cochinchinensis</i> )	(Pancapalaga et al. 2023)
		Mangrove ( <i>Ceriops tagal</i> )	(Pancapalaga et al. 2023)
	Peels	Peanut skin	(Bodoira and Maestri 2020)
		Walnut skin	(Shahidi et al. 2019)
		Canola hull	(Shahidi et al. 2019)
		Apple peel	(Shahidi et al. 2019)
		Bean seed coat	(Shahidi et al. 2019)
		Buckwheat hull	(Shahidi et al. 2019)
	Seeds	Cowpeas	(Yasmin et al. 2008)
		Grape ( <i>Vitis labrusca</i> L.)	(Smeriglio et al. 2017)
		Cacao beans ( <i>Theobroma cacao</i> L.)	(Smeriglio et al. 2017)
	Leaves	Strawberry Tree ( <i>Arbutus unedo</i> L.)	(Bule et al. 2020)
		Mangrove ( <i>Rhizophora mucronata</i> )	(Pancapalaga et al. 2023)
	Fruits	Apple ( <i>Malus domestica</i> Borkh.)	(Smeriglio et al. 2017)
		Cranberries ( <i>Vaccinium oxycoccus</i> )	(Smeriglio et al. 2017)
		Black currants ( <i>Ribes nigrum</i> L.)	(Smeriglio et al. 2017)
		Blueberries ( <i>Vaccinium myrtillus</i> L.)	(Smeriglio et al. 2017)
		Black diamond ( <i>Prunus</i> spp.)	(Smeriglio et al. 2017)
		Plums ( <i>Prunus domestica</i> L.)	(Smeriglio et al. 2017)
		Peaches ( <i>Prunus armeniaca</i> L.)	(Smeriglio et al. 2017)
		Raspberries ( <i>Rubus accidentalis</i> L.)	(Smeriglio et al. 2017)
		Pears ( <i>Pyrus communis</i> L.)	(Smeriglio et al. 2017)
Hydrolysable tannins		Bark	Sweet chestnut ( <i>Castanea sativa</i> )
	Peels	Mango ( <i>Mangifera indica</i> L.)	(Luo et al. 2014)
		Pomegranate ( <i>Punica granatum</i> )	(Elfalleh 2012)
	Seeds	Pomegranate ( <i>Punica granatum</i> )	(Elfalleh 2012)
		A mountain tea ( <i>Sideritis raseri</i> )	(Pljevljakušić et al. 2011)
	Leaves	Mango ( <i>Mangifera indica</i> L.)	(Luo et al. 2014)
		Pomegranate	(Elfalleh 2012)
		Bearberry ( <i>Arctostaphylosuva-ursi</i> L. Sprengel)	(Amarowicz and Pegg 2013)
	Fruits	Sumac ( <i>Rhus</i> spp)	(Asgarpanah and Saati 2014)
		Pomegranate ( <i>Punica granatum</i> )	(Larrosa et al. 2010)
		Raspberries	(Larrosa et al. 2010)
		Strawberries	(Larrosa et al. 2010)
		<i>Galla chinensis</i>	(Tian et al. 2009)
		Black currants ( <i>Ribes nigrum</i> L.)	(Smeriglio et al. 2017)
		Mango ( <i>Mangifera indica</i> L.)	(Smeriglio et al. 2017)
		Guava ( <i>Pisidium</i> spp.)	(Smeriglio et al. 2017)
	Raspberries ( <i>Rubus accidentalis</i> L.)	(Smeriglio et al. 2017)	
	Flowers	Pomegranate ( <i>Punica granatum</i> )	(Elfalleh 2012)





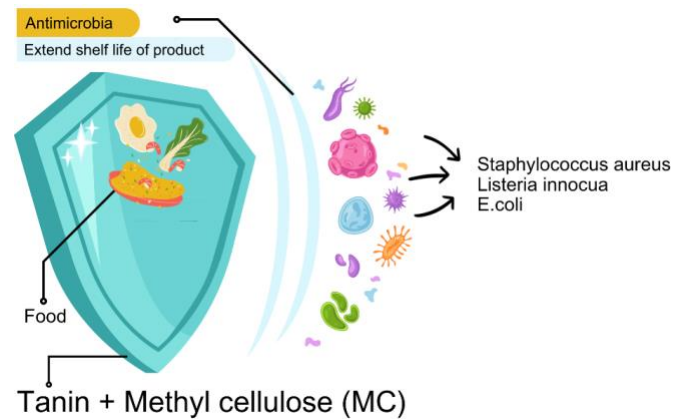
**Fig. 2.** Tannin application in the food packaging.

Several examples of active packaging, which use tannins to improve polymer properties, have recently been reported. It has been successfully demonstrated that condensed tannins can enhance the crystallization, thermal stability, and textural characteristics of polylactic acid (PLA) films (García et al. 2016). Polyvinyl alcohol containing condensed tannin has strong UV barrier properties and antioxidant activity (Zhai et al. 2018). According to Halim et al. (2018), a novel method was used to preserve grapes (*Vitis vinifera*) and cherry tomatoes (*Solanum lycopersicum* var. *cerasiforme*) for 14 days. This method involved the application of chitosan, gelatin, and methylcellulose films mixed with tannin. The study demonstrated that adding 15% tannin resulted in an augmentation of the antibacterial characteristics against *Staphylococcus aureus* and *Escherichia coli*, hence enabling tannin to serve as a food preservative. Furthermore, cellulose nanofibril films (CNFs) enhanced with tannins demonstrate exceptional antioxidant and UV-blocking properties. They can rapidly absorb 100% of light in the UV-B/UV-C range and capture over 90% of additional free radicals (Kriechbaum and Bergström 2020). Another example is a study that used tannins and poly(hydroxybutyrate-co-valerate) (PHBV) to create fully biobased and biodegradable films. The films exhibited improved gas barrier, UV protection, and antioxidant properties. Incorporating tannins into the films enhanced their thermal stability and mechanical properties, rendering them suitable for food packaging applications that range from refrigerators to high temperatures for food heating (Ferri et al. 2023).

#### 2.4.1.2. Food preservation

Contamination or attack from microbes, such as bacteria, fungi, viruses, yeast, and the like, is terrifying in food storage. All industry players need help minimizing this and chemical contamination so that food products can reach consumers. Studies show that we always use gallic acid as a standard solution in the production of sources of antioxidants and antimicrobials; unexpectedly, it is a source that comes from tannins and their groups, such as proanthocyanidins (Smeriglio et al. 2017). In one study, tannins isolated from a natural substance were described as being able to fight the bacterium *S. aureus* (Adnan et al. 2017). Then, the active compound from

the ellagitannin group, namely punicalagin, also shows antibacterial properties that have been tested for resistance to contamination from *S. aureus*, which can be utilized to reduce *S. aureus* contamination in the food industry as well (Puljula et al. 2020). In addition, the bilayer methyl cellulose (MC) film with tannin showed antimicrobial activity against *Listeria innocua* and *E. coli* (Fig. 3).



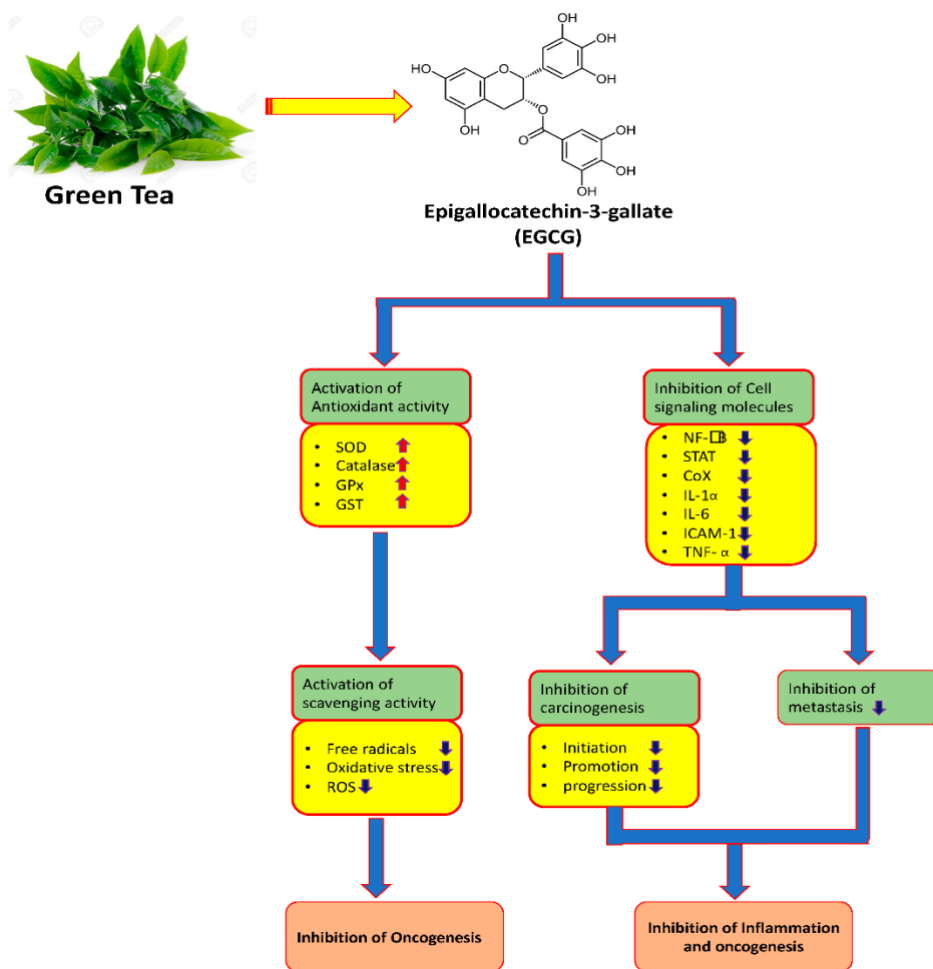
**Fig. 3.** Tannin application in food preservation.

Tannins have antibacterial activities through multiple pathways, such as enzyme inhibition, protein-cell membrane interaction for structural breakdown, and metal (iron) chelation. Tannins have an increased ability to affect gram-positive bacterial cells but can also successfully fight gram-negative bacteria, viruses, and parasites (Deshmukh and Gaikwad 2024). Commercial wood extract tannins exhibit antibacterial properties against *Salmonella typhimurium*, *E. Coli*, *S. aureus*, *Vibrio* spp., and other pathogenic bacteria (Bouarab Chibane et al. 2019). This development makes it possible to coat or package products with MC bilayer films containing tannins to prevent microbiological growth or to prolong the shelf life of goods susceptible to oxidative processes (Cano et al. 2021). Other research also states that adding tannins to biopolymer packaging increases shelf life, especially when storing climacteric fruit so that the quality is maintained and lasts longer without harming consumers (Missio et al. 2019). For example, cherry tomatoes were successfully preserved, and their quality improved for up to 20 days at room temperature when they were stored in active packaging film made of Poly Lactide-Poly (Butylene Adipate-CoTerephthalate) blends combined with tannic and gallic acid. The film demonstrated improved tensile strength, UV barrier, and antibacterial activity against *Lactobacillus monocytogenes* and *E. coli* (Sharma et al. 2022). In further research, tannin extracts derived from chestnut wood (known as tannin oenologique, TO) and grape (known as tannin VR grape, TVG) were chosen for research on maintaining the quality of minced meat during refrigerated storage. At doses of 0.25% and 0.5%, both TO and TVG are more effective than sodium metabisulfite (SMS) in preventing chemical and microbiological alterations. Therefore, these tannins could be a promising alternative to synthetic preservatives such as sodium bisulfite to control oxidation and microbial contamination (Nguyen et al. 2023).

#### 2.4.1.3. Functional food

In general, food products, an industry certainly provides a composition that follows the product's purpose. The composition itself is given in the product composition table. The

manufacturer of a food product is also prohibited from providing a product that is harmful to consumers; even in its development, a food ingredient is produced by enriching an active substance, referred to as a functional food or nutraceutical product. One of the active substances included in food is tannin. Tannins in functional foods are very beneficial because they have functional properties that can prevent diseases such as diabetes, heart disease, kidney disease, cancer and other degenerative diseases (Teodor et al. 2020). The sources of natural tannins already available in nature have been produced from tea leaves, apples, chocolate, grapes, cherries, and other berries. Green tea contains condensed tannins, which can provide anti-cancer activity due to its anti-inflammatory and antioxidant properties (Fig. 4) (Ratnani and Malik 2022). As for other sources, epigallocatechin gallate can also inhibit the Cyclooxygenase-2 (COX-2) enzyme, which leads to the synthesis of prostaglandins in several types of colon cancer cell lines. As a further explanation, epigallocatechin gallate is a potent antioxidant that can prevent and reduce the activity of nitric oxide and malonaldehyde free radicals and increase the enzyme superoxide dismutase in the colon (Liu et al. 2019; Pratama et al. 2022a).



**Fig. 4.** Green tea shows anti-cancer activity due to its anti-inflammatory and antioxidant properties (Huang et al. 2020).

#### 2.4.2. Wood composites industry

The wood composite industry plays an important role in life, mainly because basic human needs are food, clothing, and shelter. The timber industry enters the housing realm, an inseparable part of the line of life. Wood adhesives are crucial for assembling and bonding wood components



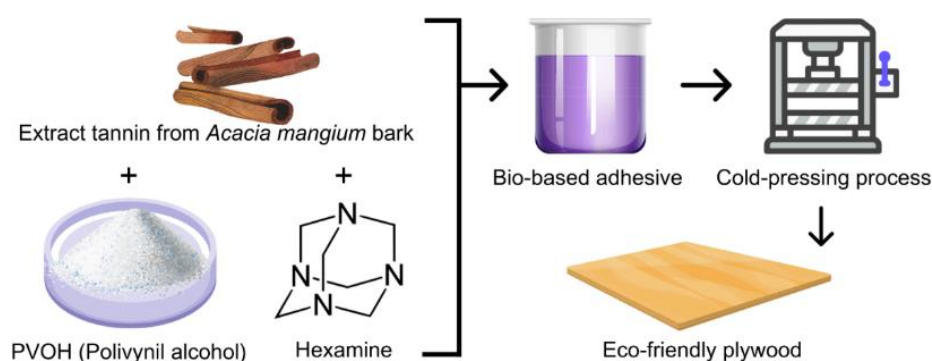
to produce wood-based panel products. The adhesive's quality significantly impacts the performance of the final wood product, making it a crucial factor in wood processing. Formaldehyde resins containing urea, phenol, and melamine are the most widely used adhesives. Formaldehyde is the fundamental component in approximately 95% of the wood adhesives employed in producing engineered wood composites (Kumar and Pizzi 2019). The predominant resin type in global resin production is urea-formaldehyde (UF), accounting for approximately 85% of the overall resin market. Melamine follows in second with 10%, followed by phenolics with 5% (Costa et al. 2013; Kumar and Pizzi 2019; Park and Kim 2008). The advantages of UF adhesives include their cost-effectiveness, non-flammability, rapid curing time, and light coloration. Nevertheless, it is important to note that UF resins containing formaldehyde have been found to have adverse effects on human health. Furthermore, polyurethanes (PU) have garnered extensive utilization as adhesives for wood materials. Isocyanate-based PU demonstrates exceptional robust adhesion, flexibility, and durability (Aristri et al. 2023). However, it possesses limited renewability and is associated with toxicity concerns. Therefore, it is imperative to identify a viable substitute for producing PU resins and adhesives that do not include formaldehyde.

There has been an increasing interest in developing adhesives from plant sources, specifically tannins, due to their environmental benefits and ability to improve performance. Using tannin-based wood adhesive presents several notable benefits, including its environmentally sustainable nature, non-toxic properties, and commendable adhesive strength. Virginia et al. (2020) research showed that the rind of Salak fruit, which contains tannins, was used to create a tannin-based adhesive. The extract was reactive to formaldehyde, and the adhesive was tested for its bonding strength. The results indicated that using tannin-based adhesive from Salak fruit was a potential alternative raw material for bio-adhesives for laminated wood. Tannins from *Myrcia splendens* were used to formulate a urea-tannin-formaldehyde (UTF) resin for application to glue lines in *Pinus* sp. Incorporating tannins into the adhesive enhanced the strength of the glue line. According to Danielli et al. (2021), the optimal proportion for the formulation was 5.0% tannins. Then, a study demonstrated the synthesis of wood glue that is entirely bio-sourced through a covalent reaction between quebracho tannin, a commercial flavonoid tannin, and soybean protein isolate (SPI). The adhesive was non-toxic and showed good plywood bonding results when reacted at 40°C and then hot pressed at a higher temperature (Ghahri et al. 2022).

The development of PU adhesives from hydrolyzed or condensed tannins can be categorized into two primary techniques. The first involves modifying the tannins in flavonoid compounds to promote reactions with isocyanates. The second technique employs a non-isocyanate methodology to guarantee the ecologically sustainable nature of the developed adhesive (Aristri et al. 2021). Condensed tannins were employed to develop non-isocyanate polyurethane resins (NIPU) based on tannins. These resins undergo a reaction with dimethyl carbonate (Thébault et al. 2015). Then, hexamethylenediamine is added to this mixture to create a urethane bond. Particleboard and medium-density fiberboard can be produced using NIPUs derived from tannins (Hemmilä et al. 2017). In Aristri et al. (2023) research on ramie fiber, NIPU resin was utilized to modify ramie fiber, namely fiber derived from the stems of the ramie plant (*Boehmeria nivea* L.) using tannin-based non-isocyanate polyurethane (tannin-Bio-NIPU) and tannin-based polyurethane resin (tannin-Bio-PU). Tannin extract, dimethyl carbonate, and hexamethylene diamine combine to form tannin-Bio-NIPU resin, whereas tannin-Bio-PU is synthesized using diphenylmethane diisocyanate (pMDI) polymer. Tannin-based NIPU has several benefits, including enhanced strength, reduced curing temperature, decreased pressing time, and the absence of isocyanates. The

latest advancements in tannin-based NIPU resins in wood adhesives exhibit potential as a sustainable and ecologically sound substitute for isocyanate-based PU.

In recent years, there has been a growing emphasis on acquiring knowledge about environmental protection and personal wellness. As a result, there has been significant interest in natural resins derived predominantly from renewable sources. This interest stems from the phenolic composition of tannic resins, which enables their application as adhesives and as a potential substitute for phenol in adhesive formulations. The study and implementation of resin have yielded significant achievements in numerous nations (Zhou and Du 2020). The reaction of tannins is primarily driven by their phenolic nature, which is comparable to that of phenols with formaldehyde, owing to their structural resemblance to synthetic phenols, as noted in reference (Ping et al. 2011). Tannins have been identified as a prospective alternative to phenol in the composition of phenol-formaldehyde resins for wood adhesive formulations. The results indicate that it is possible to manufacture exterior-grade plywood using by-products from the forestry sector without using phenols, formaldehyde, or isocyanates (Aristri et al. 2022). Lignin and tannins have been used to form diverse wood adhesives to produce particleboard and plywood that meet relevant interior standards (Aristri et al. 2021). A diverse range of biologically derived compounds can be effectively integrated with tannins to produce sticky materials. The present study has formulated an adhesive composed of maize starch, mimosa tannin, and hexamine as a hardening agent. The adhesive used for cold-setting plywood is formulated using a combination of PVOH (polyvinyl alcohol), tannin, and hexamine, commonly called PTH adhesive. Sari et al. (2023) investigated the impact of adhesive composition, namely varying concentrations of tannin and hexamine, as well as different durations of cold pressing (3, 6, 12, and 24 hours), on plywood's physical and mechanical properties (Fig. 5). The findings of this study indicate that the manipulation of tannin and hexamine levels, as well as the duration of cold pressing, can enable the production of plywood according to the strength criteria set by Japanese standards. This study investigates the viability of utilizing cold-pressed eco-friendly plywood panels bonded with PTH-based glue as a potential substitute for conventional plywood.

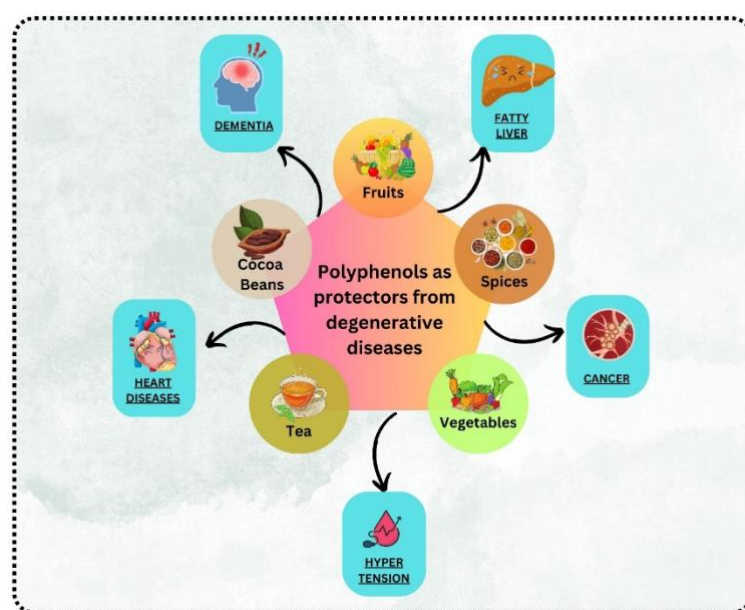


**Fig. 5.** PTH-based glue for conventional plywood.

#### 2.4.3. Medicines

Since the Industrial Revolution, the development of drugs since the industrial revolution has created many synthetic drugs that are pretty capable of incriminating the human body and having adverse effects in the future (Brunt et al. 2017). So, the researchers created and tried to find drugs that are more natural and able to become alternative medicines to their predecessors. The

polyphenol group is a secondary metabolite compound from plants with functional properties as an antioxidant because it has a -OH hydroxyl group that can tame free radical compounds to stop the oxidation chain reaction in the body (Pratama et al. 2023; Abbas et al. 2017). This performance can be linked to preventing degenerative diseases such as heart disease, diabetes, liver damage, tumors, cancer, and the like (Fig. 6) (Brglez Mojzer et al. 2016). Most of the chemical products based on tannins, such as gallic acid, gallic epigallocatechin, and proanthocyanidins, have been tested and found to be very effective antioxidants. Then, the inclusion of tannins into the alcohol group performs tannins in their antibacterial properties, such as preventing damage caused by the pathogenic bacteria *Staphylococcus aureus* and pathogenic microorganisms that tend to be resistant to several drugs which are a health problem in the human population (Okuda and Ito 2011).



**Fig. 6.** Health benefits effect of polyphenols.

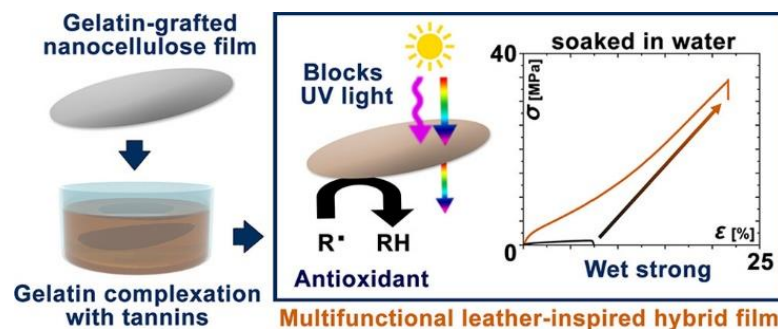
### 3. Tannins-Based Hybrid Materials

Tannins are a group of compounds in which molecules have the advantage of combining with other molecules. The bonds formed by the interaction of hydrophilic and nucleophilic groups with polymers or polymer surfaces, for example, biopolymers, microfibers, nanofibers, and synthetic polymers, which will form a new product whose physical and chemical properties usually increase and increase in functional properties. However, few functional properties appear and provide a slight opposite change from the nature of the natural polymer. So, we need a model that continues to be studied to get the optimal value of the new product for the functional properties it produces. The tannin-based polymers studied for several years are as in the following sub-chapter.

#### 3.1. Tannin-Based Nanocomposite Products

The old composite product is usually made with the basic ingredients of fiber components or fibers, which are then combined with tannins. However, many breakthroughs have been made, and composites with smaller sizes have been created through a more even mixing process. This process will produce small products, such as nanoflakes, nano and microfibers, or carbon

nanotubes (CNT). Usually, the obstacles or factors for mixing are the homogenization process, the length of time, solvents, and the ratio of solvents to materials used (Saldanha et al. 2018; Wang et al. 2017). This packaging, which is a combination of nanofibers and tannins, has the advantage that it can be used to manufacture food packaging products (Missio et al., 2018). CNTs with robust applications can be used as polymers for heavy equipment or automotive, aerospace, and aeronautics (Sivasankaran et al. 2017). The character of the fiber material hybridized with tannins is that it can withstand chemical and physical damage, such as oxidation damage, due to the antioxidant properties of the tannin compound and can protect the material from the sun's UV rays. A study demonstrated a leather-inspired method to create versatile materials with strong moisture resistance. This method involves using tannin to deposit gelatin onto cellulose nanofibrils (CNF) that have been changed on the surface (Fig. 7). This film has strong antioxidant and UV-blocking characteristics by rapidly scavenging over 90% of extra free radicals and absorbing 100% UV-B/UV-C radiation (Kriechbaum and Bergström 2020). Regarding its application as food product packaging, the composite product's phenolic properties may also confer antimicrobial properties (Li et al. 2019).



**Fig. 7.** Leather-inspired approach to produce multifunctional film (Kriechbaum and Bergström 2020).

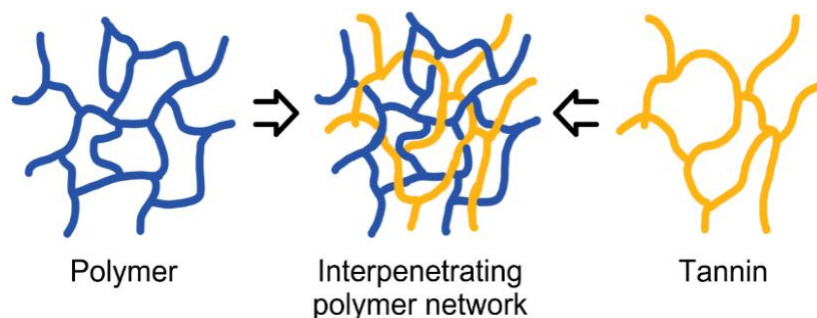
Then, in its development, polysaccharide compounds also need to be oxidized to aldehyde compounds to maximize the combination with tannin compounds (Ji et al. 2020). Previous studies showed that covalent bonds occur with tannic acid in epoxidized soybean oil in the manufacture of cellulose microfibrils because the epoxy group easily reacts with phenolic alcohols, causing the network to become stronger. Thus, tannic acid functions as a reinforcing agent that can accelerate the process of making composites from cellulose, which are usually challenging to disperse to form a mixture (Ji et al. 2020).

### 3.2. Interpenetrated Network Products and Polymer Blends

In addition to what has been described in the previous point, the manufacture of composites with polymer mixtures can also be carried out without tube cores, fibers, or debris. Composites can be made directly by mixing with tannic acid from natural polymers. One example is natural rubber directly mixed with tannic acid, which can increase the strength of the polymer formed by two hundred percent, much higher than commercial vulcanized rubber products (Yang et al. 2017). The interpenetration network (Fig. 8), also described in several sources, can increase the strength and resilience of a composite because it can increase the product's flexibility (Kadokawa et al. 2011). This is also evidenced in composite products with nano sizes derived from natural



polymers, namely poly-lactic acid or PLA, and polyurethane networks formed from propylene oxide tannins obtained from polymethyldiisocyanide reactions (García et al. 2018).



**Fig. 8.** Interpenetrating networks.

Besides increasing the mechanical ability of composites produced from natural products, tannins can also increase the chemical and biological capabilities of the resulting products. The phenolic hydroxyl group tannins possess means that mixing polymers with tannins can ward off free radicals such as reactive oxygen species (ROS). The effect of condensed tannins derived from larch bark and polyvinyl alcohol can provide an antioxidant effect on the resulting product (Zhai et al. 2018). The ethylcellulose film product, produced and hybridized with tannins derived from grape seed, also showed its antioxidant properties (Olejar et al. 2014). However, the mechanical value of the resulting composite may be sacrificed when mixing natural polymers with tannins, mainly if the reaction cannot produce covalent bonds. This is exemplified in the manufacture of chitosan polymer with tannic acid (Rubentheren et al. 2015). Then, when tannins interact with water in their application, this interpenetration network can reduce the critical temperature, making tannins easily soluble in an aqueous environment. It is applied to polyvinyl-caprolactam with tannins (Costa et al. 2011).

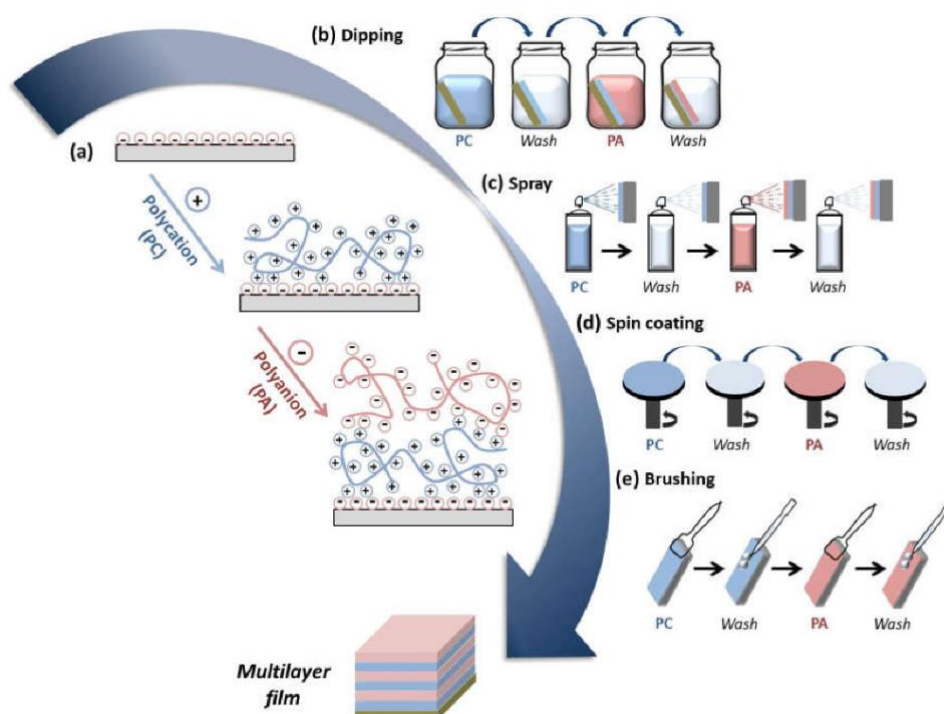
### 3.3. Polyelectrolyte Complex Products, Layer by Layer like Materials and Coatings

Polyelectrolyte complexes are association complexes formed between polyions with opposite charges due to electrostatic interactions between these charged polyions. Usually, this is done by combining polymers with other polymers. The aim is to produce polymer products with improved physical and chemical properties. Hydrogen bonds and hydrophobic interactions between tannins and gelatin can induce coacervation. Therefore, hydrolyzed tannins can make nanospheres in a low concentration range to speed up forming a stable suspension (Jridi et al. 2019). As for this product and related research, combining epigallocatechin-3-gallate with tannins derived from bayberry can absorb toxic  $Hg^{2+}$  ions. The resulting product can be appropriately used and economical (Wu et al. 2009). Similar results were found in the investigation of electrospun nanofiber membranes constructed from a blend of polyvinyl alcohol and gelatin with tannins, which improved their physical properties and had high uranium adsorption capacities in a seawater solution system (Meng et al. 2019).

The production of polyelectrolyte complexes (PECs), composed of natural polycations and polyanions through electrostatic interactions, is one method for improving the biostability of natural polymers under physiological conditions without compromising their biocompatibility. Through the alternating deposition of oppositely charged molecules, multilayer assembly using a layer-by-layer (LbL) approach based on electrostatic interactions yields multilayer films with well-



controlled structure, composition, and thickness. Since many natural polymers in this situation have ionizable groups in their chemical structure, they are polyelectrolytes. This means that electrostatic interactions between positively charged polymers (polycations) and negatively charged polymers (polyanions) can occur through the LbL technique (**Fig. 9a**) (Criado-Gonzalez et al. 2021). Dipping, spraying, and spin coating are the most widely used LbL techniques. The dipping technique is carried out by immersing the substrate alternately in a polycation and polyanion solution with a washing stage between the deposited layers (**Fig. 9b**) (von Klitzing et al. 2012). The spraying technique is carried out by sequentially spraying polycation and polyanion solutions to produce a multilayer film in a few seconds (**Fig. 9c**) (Behler et al. 2009; Izquierdo et al. 2005). The spin coating technique is carried out by spinning the substrate so that polymer deposition occurs (**Fig. 9d**) (Maziukiewicz et al. 2020). Apart from that, there is a brushing technique, namely brushing a polyelectrolyte solution sequentially over the substrate (**Fig. 9e**) (Park et al. 2018).



**Fig. 9.** Scheme of a layer-by-layer (LbL) assembly of polyelectrolyte; several techniques to build multilayer films: (a) Multilayer film; (b) Dipping; (c) Spray; (d) Spin coating; and (e) Brushing (Criado-Gonzalez et al. 2021).

Then, layer by layer, products are used for products that require properties to resist microbiologic damage. This is explained by the fact that collagen products bound with tannins can provide stability against enzymatic degradation through the inactivation mechanism of oxidizing enzymes (Natarajan et al. 2013). This can also be used in the biomedical field. In addition, its adhesive capacity with bactericidal properties can be exploited in forming tannic acid products with gelatin and further cross-linking the modified silver nitrate biopolymer (Guo et al. 2018). Another example is persimmon tannin, which can bind covalently to chitosan through glutaraldehyde. This can produce a full biopolymer-based combined product that significantly

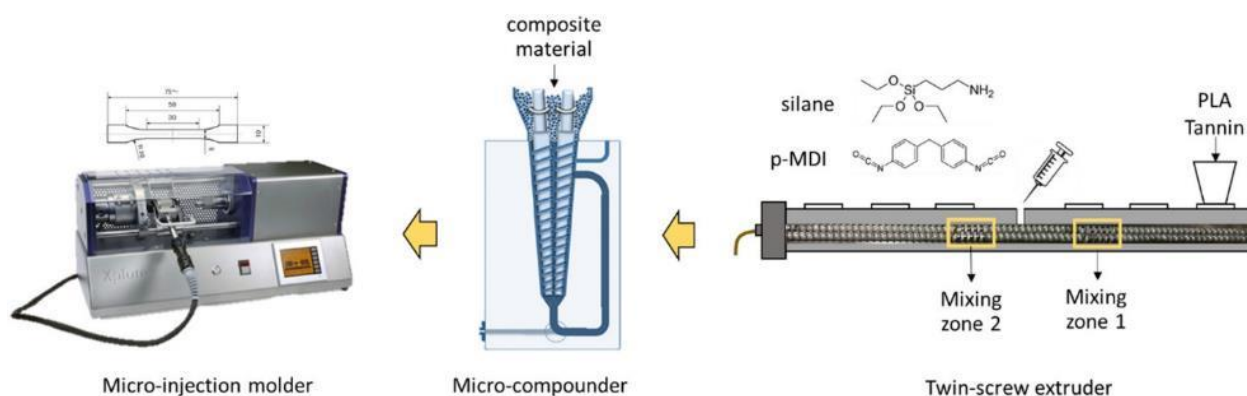
increases the ability to efficiently and selectively adsorb toxic  $\text{Pd}^{2+}$  in wastewater, usually rhodamine dye wastewater (Zhou et al. 2015).

In products where tannins act as coatings, tannins are combined with polymer compounds or metals used as facilities, provided they are resistant to physical and chemical qualities and can avoid microbial attacks. This is exemplified in the conductive hydrogel used in spinal treatment (Zhou et al. 2018). Another explanation relates to polypyrrole polymers, which bind to tannic acid and Fe(III) ions, in which tannic acid can act as a physical, chemical, and biological oxidizing agent (Chen et al. 2019). This combination has a positive effect on the treatment of spinal cord injury, which has been tested in vivo. The use of tannins with biopolymers has been extensively tested in recent studies. The nature of tannins can support the biopolymer in which it is sheltered so that its quality also increases. So far, there is no doubt that tannins' wide availability and usefulness can be utilized.

#### 4. Method of Combining Tannins with Biopolymers

##### 4.1. Preparation of PLA-Tannin Biopolymer with the One-Step Reactive Extrusion Method

Poly(lactic acid), or PLA, is one of the most widely used and safe biopolymers in food packaging. PLA has desirable qualities such as strong mechanical properties, good appearance, and low toxicity. The drawback of PLA is that it is highly permeable to gases and vapors, which raises the possibility of oxidation in food packaging. This can solve this weakness of PLA if combined with tannin active compounds, which can be free radical scavengers. However, developing PLA-tannin biocomposites with increased properties is difficult due to the compatibility issue between PLA and tannins. One-step reactive extrusion method with the help of 3-aminopropyltriethoxysilane (APS) or methylene diphenyl diisocyanate (p-MDI) can produce PLA-tannin biocomposites with good properties (Fig. 10) (Liao et al. 2020).



**Fig. 10.** Reactive extrusion method using a twin-screw extruder (Liao et al. 2020).

The one-step compatibilization method is a process of mixing with the help of adhesion agents (e.g., compatibilizers and support agents) to develop composites with good compatibility. As a result, this approach may enhance the adherence of the component interface and, consequently, thus filler distribution into the polymer matrix during the extrusion process. (González-López et al. 2018; Nyambo et al. 2011).

The composite was evaluated by morphological, rheological, tensile, and thermal properties to see the level of compatibility produced through the one-step compatibilization method. The

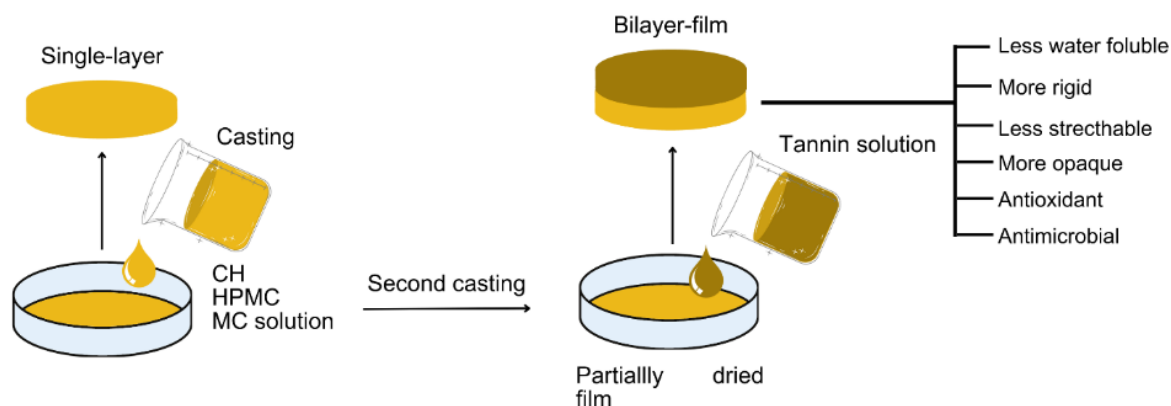
results show that p-MDI (methylene diphenyl diisocyanate) can efficiently improve the tensile behavior of composites with strong tensile fracture surfaces compared to incompatible PLA/tannin composites. In addition, these composites provide enhanced viscosity properties, plateau rheological behaviors, thermal degradation, and high melting temperatures (Liao et al. 2020).

Cross-links are formed between tannins and p-MDI/APS during the mixing process, leading to rigid thermoset particles dispersed into the PLA matrix as internal cross-linked domains (Lu et al. 2022). These particles have various reactive functional groups to further act as local cross-linking points to link the PLA chains (Tee et al. 2017). Consequently, the interaction between PLA/tannin and p-MDI/APS leads to complex interactions, enhancing the adhesion at the interface and promoting stress transfer between the two phases. As a result, the tensile strength is significantly higher compared to the incompatible combination of PLA and tannin. Thus, this method can be applied efficiently to develop PLA-based biocomposites or other biopolymers (Liao et al. 2020).

#### 4.2. Preparation of Cellulose-Tannin Biopolymer with Layer-by-Layer Method

Developing edible coatings and biodegradable films that act as transporters for active compounds like polyphenols is now being studied using several polysaccharides, including chitosan, methylcellulose, and hydroxypropyl methylcellulose. To enhance the antimicrobial and antioxidant properties, essential oils, olive leaf, green tea or pomegranate seed extract, or resveratrol are added to the cellulose and chitosan-based films (Nemazifard et al. 2017; Sánchez-González et al. 2010; Siripatrawan and Harte 2010).

In the study of Cano et al. (2021), the development of bioactive polymers used different polysaccharides, namely chitosan (CH), hydroxypropylmethylcellulose (HPMC), and methylcellulose (MC), with the addition of tannin bioactive compounds from three different sources, namely oak bark, grape skin red, and white grape skin. Films enriched with tannins are obtained by casting using the single-layer method (mixing all components) and the bilayer method (one layer of polysaccharide plus one layer of tannin as coating). The monolayer was prepared by mixing tannin into the polysaccharide film-forming solution and stirring continuously until homogeneous. Then, double layers were prepared by pouring aqueous tannin solutions onto partially dried carbohydrate films for 24 hours (Fig. 11).



**Fig. 11.** Preparation of bilayer film with layer-by-layer method.

Based on microscopic analysis, the films produced with monolayer composites showed a more heterogeneous cross-sectional layer, indicated by the tannins partially mixed with the

polymer. In the bilayer film, two distinct layers are observed: the upper layer, which is rich in tannins, and the lower layer, which has little tannins. This is due to a sudden concentration gradient at the interface, as expected from a preparation method in which a tannin solution is added to a partially dried polymer film. The tannin layer on the top looks compact but shows some cracks due to its brittleness without a polymer ratio that is high enough. In contrast, each bilayer film has a surface that appears cracked because the tannin layer is so brittle after drying (Pinotti et al. 2007).

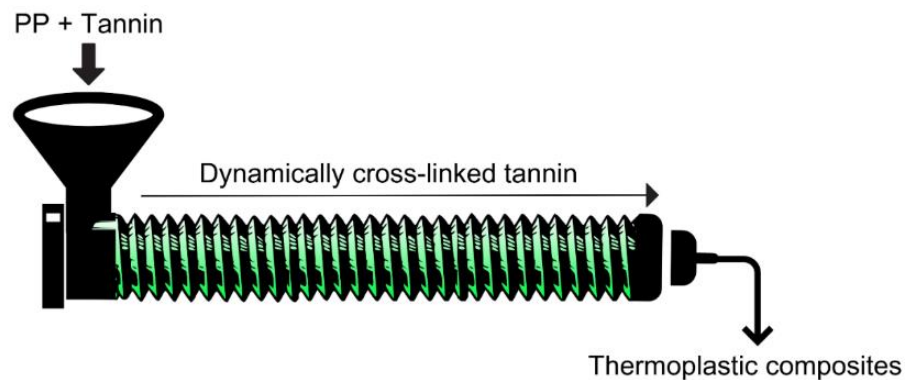
Several polysaccharides, namely CH and MC, bind polyphenols well because of the hydrophobic and hydrogen bonding interactions (Yu et al. 2015). Tannins serve as binding agents in these composites, which results in a less soluble in water, stiffer, less stretchy, more opaque, and slightly less glossy film. Based on the method, the bilayer film has a higher antioxidant content because the tannin content is more on the film's surface. However, of the three polymers used, MC bilayer films with tannins produce the best film packaging to prevent oxidative processes or control bacterial growth to extend product shelf life (Cano et al. 2021).

In addition, CMC and PLA are known as environmentally friendly polymers that can be made into composites for food packaging applications (More et al. 2023). Nevertheless, the inherent interfacial differences between CMC and PLA pose challenges and maybe layer separation problems (Liao et al. 2021). Xiao et al. (2024) introduced a direct approach to mitigate these challenges by incorporating tannic acid and ferric chloride in CMC-PLA preparation. The formation of a cohesive interface improves interlayer compatibility. CMC/TA-PLA multilayer films were prepared by mixing CMC and TA in distilled water and then casting them into films. Next, PLA was dissolved in a dichloromethane solution, and FeCl<sub>3</sub> was added. The CMC/TA film was immersed in this solution for 10 seconds and allowed to evaporate. Without any layer separation, the resulting CMC/TA-PLA/Fe multilayer film exhibits excellent mechanical strength, high contact angle, and superior thermal stability. In addition, the CMC/TA-PLA/Fe film showed excellent efficacy in blocking ultraviolet rays, effectively minimizing discoloration on various wood surfaces subjected to UV aging.

#### 4.3. Lignin-Tannin Biopolymer Preparation with Dynamic Vulcanization Method

Polypropylene (PP) is a thermoplastic formed from propylene monomer and has good mechanical properties, low density, and low cost, making it the most popular commodity (Maddah 2016). Over the last decades, PP has been combined with natural polymers to reduce the use of petroleum-based plastics (Visakh and Poletto 2017). In addition, PP also has properties that are very sensitive to oxygen and UV light compared to other plastics, so it can be combined with natural biopolymers, such as tannins and lignin, to overcome these problems. This is because the structure of the tannin and lignin flavonoids can prevent photo and thermo-oxidative degradation by stabilizing itself into a stable phenoxy radical and preventing the formation of new radicals (Tolinski 2009).

The dynamic vulcanization method is one method of mixing tannin/lignin with PP. The dynamic vulcanization method is a traditional technique for preparing thermoplastic-elastomer composites by extrusion (Fig. 12). During vulcanization, high melting temperature and thermoplastic shear rate are required to trigger cross-linking between the elastomer and the thermoplastic polymer during the mixing process (George et al. 2000). Several studies have successfully used dynamic vulcanization methods for PP/thermoset resins, such as epoxy and novolac (Cui et al. 2007; Liao et al. 2019; Nakason et al. 2008).



**Fig. 12.** Dynamic vulcanization process for preparing thermoplastic composites, adapted from Liao et al. (2019).

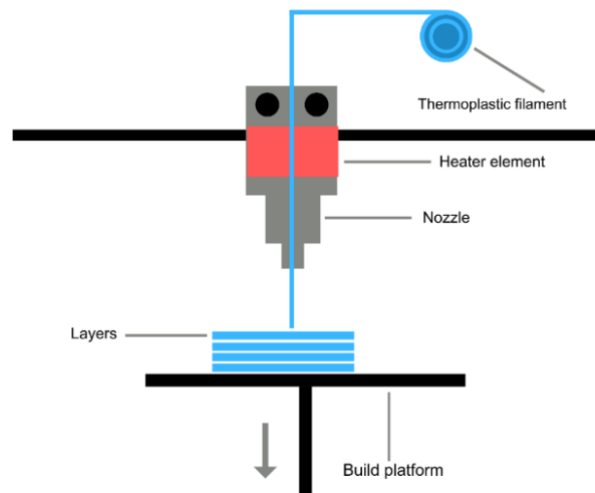
The dynamic vulcanization method has been successfully applied to mixed PP and tannin/lignin composites. The resulting PP-tannin/lignin composite has higher thermal stability, modulus, and stiffness characteristics than PP without a mixture. The increase in Young's Modulus in this composite occurs due to the extrusion process, which causes the cross-linking of tannins and lignin to become rigid thermoset particles. After the extrusion process, the hydrophilicity of tannin/lignin decreased, resulting in increased compatibility with PP. In addition, vulcanized tannin/lignin can act as a nucleating agent for PP and has good thermal stability compared to tannin/lignin alone. Based on rheological data, vulcanized tannin/lignin can be dispersed better into the PP matrix than native tannin/lignin. In addition, the resulting composite has good anti-UV properties, characterized by small changes in surface morphology, tensile properties, crystallinity, viscosity, and carbonyl index (Liao et al. 2021).

#### 4.4. PLA-Tannin Biopolymer Preparation by Acetylation Method

PLA is one of the most promising biopolymers because it has several unique properties, especially its biodegradability, biocompatibility, and thermoplastic processability. PLA can be used as a 3D printed material such as plastic cups, plastic bottles, and medical implants using a material extrusion process technology known as fused deposition modeling (FDM). The principle of the FDM method is that a thermoplastic filament is inserted into the nozzle channel, which is driven by a motor. The nozzle heats the filament, and then the melted part comes out of the nozzle tip to form layer-by-layer 3D printing material (Fig. 13).

Biomaterials or biopolymers hold great promise as raw materials for 3D printing due to their biodegradability, sustainability, harmlessness, and cost-effectiveness. The manufacture of PLA composites combined with tannins is being studied through 3D printing. The main objectives of this study are to devise a novel technique for incorporating tannins into PLA for use in 3D printing constructs and to evaluate the feasibility of FDM. However, the dispersion of tannins into PLA can cause nozzle clogging and deposition, which is a challenge in 3D printing. To overcome this challenge, tannins are processed by acetylation with acetic anhydride to reduce their hydrophilic properties so that the compatibility of tannins with the PLA matrix increases (Grigsby et al. 2013). The process of making PLA-tannin composite filaments was carried out using a twin screw extruder. The resulting PLA-tannin acetate (AT) composite filaments will be evaluated for color change, crystal properties, thermal degradation, hydrolysis degradation, and the 3D printing process capability for the PLA-AT composite.





**Fig. 13.** The Fused Deposition Modelling (FDM).

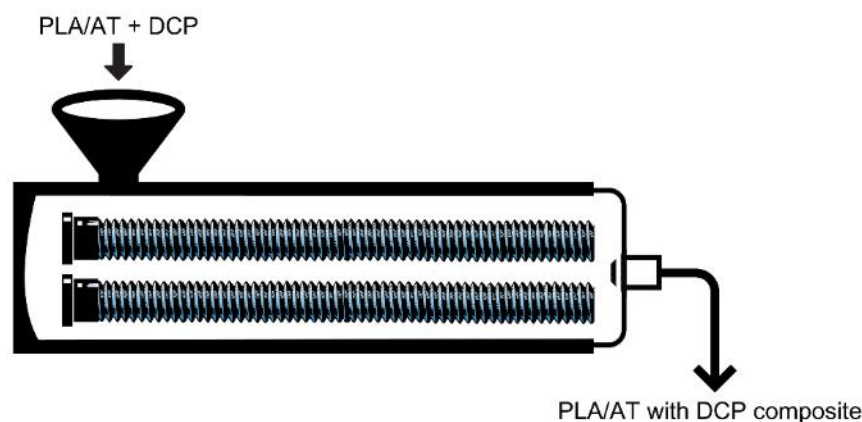
PLA combined with acetylated tannin (AT) did not significantly affect the final product's surface morphology and mechanical properties. The use of AT in PLA did not have a significant effect on the melting temperature and glass transition. Based on the thermogravimetric analysis, low temperatures below 220°C in the printing process produce a good final product without causing material decomposition. When using high temperatures in the printing process, phase separation and AT aggregation will occur, particularly on filaments with high AT levels, which can cause print defects in the final product. Under alkaline conditions, PLA-AT composites have higher hydrolytic degradation properties than PLA without AT, so that these properties can be applied in biomedical fields such as implant devices (Liao et al. 2020).

#### 4.5. Preparation of PLA-Tannin Biopolymer with In-Situ Extrusion Method

Recently, chemical modification of biopolymers using coordinating agents or free radical grafting approaches can improve the compatibility between hydrophilic and hydrophobic biopolymer matrices. The free radical grafting method is a simple technique that can improve the compatibility of multiphase composites due to increased composite interface adhesion and phase stress transmission resulting in good mechanical properties (Ma et al. 2014; Semba et al. 2006; Wang et al. 2014; Wei and McDonald 2015). Peroxide initiators can generate free radicals during the smelting mixing process, inducing chain cleavage, branching, and cross-linking. One example of a peroxide initiator is dicumyl peroxide (DCP). Dicumyl peroxide (DCP) has been successfully used as a peroxide initiator to produce bacterial polyester poly(3-hydroxybutyrate-co-3-hydroxyvalerate) and Kraft softwood composites by increasing interfacial adhesion during the melt extrusion process resulting in good mechanical, rheological, thermal properties (Luo et al. 2016). However, based on previous studies, incorporating tannin into PLA using peroxide initiators resulted in very brittle composites. Thus, in the study of Liao et al. (2021), the reactivity of tannins was modified by esterification using acetic anhydride to modulate the kinetics of complex reactions during the extrusion process between DCP as the initiator of peroxide, tannin acetate (AT), and the PLA matrix.

In-situ free radical grafting using a DCP initiator increased the adhesion of PLA and acetate tannin (AT) interfaces. The mixing process of PLA and AT is carried out in a twin screw extruder (Fig. 14). The copolymerization reactions that occur are complex, including chain-cutting,

grafting, and branching reactions from the PLA/AT matrix mixture. The PLA/AT mixture modified with a DCP initiator yields a polymer with good Tensile strength and Young's modulus properties, exhibiting strong filler/matrix interfacial strength. The increase in molecular weight of the PLA/AT mixture indicates that the cross-linked structure is formed through a chain grafting or branching reaction between PLA and AT. The cross-linked structure of the PLA/AT mixture formed also causes a higher glass transition temperature ( $T_g$ ) because the mobility of the polymer chain macromolecules becomes more difficult. Compared to PLD (PLA coated with DCP as a control), the cross-linked structure of PLA/AT blends produces higher complex viscosities, storage moduli, and loss moduli. Based on the research results, a high content of tannin acetylation results in higher grafting efficiency. This in-situ reactive extrusion method with graft modification can be applied to develop thermoplastic/biopolymer blends with tannins to produce value-added polymers with active properties as packaging and reduce material costs (Liao et al. 2021).



**Fig. 14.** Preparation PLA/AT with DCP composite using a twin screw extruder, adapted from Harnkarnsujarit et al. (2021).

## 5. Mechanism of Tannin as an Active Packaging

Tannins are natural polyphenolic compounds in various plants, especially fruit seeds, skin, leaves, and roots. Tannins can be used as an active packaging material in various applications in the food industry. The following are some of the main mechanisms that explain how tannins can function as active packaging:

### 5.1. Antioxidant Properties

Tannins can be incorporated into packaging materials to increase antioxidant properties and help protect packaged foods or beverages from oxidative damage. Tannins have strong antioxidant properties. They can protect food and beverage products from oxidative damage from exposure to oxygen and light. Thus, tannins can help maintain product quality for longer. The mechanism of tannin as an antioxidant in packaging materials involves its ability to capture free radicals and chelate metal ions, which can catalyze oxidative reactions (Ferri et al. 2023; Missio et al. 2020). Tannin is a polyphenolic compound containing hydroxyl groups, which can donate hydrogen atoms to free radicals to neutralize them and prevent oxidative damage to packaged food or drinks. Furthermore, tannins can form complexes with metal ions like iron and copper, facilitating the generation of free radicals via the Fenton and Haber-Weiss reactions. Through chelating these metal ions, tannins can decrease lipid peroxidation, the primary source of oxidative damage in

food and beverage products, and prevent the production of free radicals. (Moccia et al. 2020b; Motta et al. 2020; Pratama et al. 2021). The antioxidant properties of tannins have been demonstrated in various studies, such as tannins from *Acacia mearnsii* have been used in the development of functional hybrid films with cellulose nanofibrils (CNF), producing materials with high and prolonged antioxidant effects and increased hydrophobicity (Missio et al. 2020). Additionally, tannins have been incorporated into poly(hydroxybutyrate-co-valerate) (PHBV) films for food packaging, enhancing antioxidant, UV protection, and gas barrier properties, with the best combination observed at 5 per hundred resin tannin (phr) content (Ferri et al. 2023). Tannins have also been used in other applications, such as guar gum films as colorimetric pH indicators with antioxidant and antimicrobial potential (Emir 2023). Additionally, tannins have been used to develop antioxidant coatings with superior properties, such as enhanced UV-C irradiation protection on polystyrene films (Oliveira et al. 2023).

## 5.2. Antimicrobial Properties

Tannins inhibit the growth of microorganisms through several mechanisms, making them useful in packaging materials for extending the shelf life of food and beverage products. The specific antimicrobial properties of tannins that contribute to their effectiveness in inhibiting microbial growth include:

- Interference with enzyme activity: tannins can interfere with the activity of microbial enzymes, disrupting vital metabolic pathways and cellular functions. This interference can lead to the inhibition of microbial growth and survival (Nsahlai et al. 2011).
- Cell membrane disruption: tannins can damage microorganisms' cell membranes, releasing cellular contents and ultimately leading to cell death. This disruptive effect on microbial membranes contributes to the antimicrobial activity of tannins (Othman et al. 2019).
- Chelation of metal ions: tannins can chelate metal ions, which are essential for the growth and survival of microorganisms. By chelating these ions, tannins can interfere with microbial metabolic processes, inhibiting their growth and proliferation (Liu et al. 2021).
- Precipitation of proteins: tannins can precipitate microbial proteins, which can disrupt essential cellular processes and contribute to the inhibition of microbial growth (Shathi et al. 2022)
- Dose-dependent inhibition: the antimicrobial effect of tannins is often dose-dependent, meaning that higher concentrations of tannins result in greater inhibition of microbial growth. This dose-dependent inhibition makes tannins effective at controlling microbial proliferation in packaged products (Othman et al. 2019).

Several studies have investigated the antimicrobial characteristics of tannins and their possible uses in packaging materials. Tannins from the leaves of *Vitex doniana* against various microbial isolates, including *Escherichia coli*, *Staphylococcus aureus*, *Salmonella typhi*, *Pseudomonas aeruginosa*, and *Candida albicans* with a dose-dependent inhibitory effect against the microbial isolates (Njokuocha and Ewenike 2020). Another study investigated the antimicrobial properties of tannins from *Phyllanthus columnaris* stem bark against Gram-positive cariogenic bacteria, obligate anaerobic Gram-negative periodontopathic bacteria, and *Candida* species. The tannins inhibited the growth of all tested pathogens at minimum inhibitory concentrations ranging from 0.16 to 1.25 mg/mL, demonstrating their broad-spectrum antimicrobial activity (Othman et al. 2019). Additionally, research on pomegranate extracts from the peel and male flower by-products (Gigliobianco et al. 2022), *Phyllanthus columnaris* stem bark extract (Othman et al. 2019), conifer leaves extract (Ushio et al. 2013), demonstrated the

antimicrobial activity of tannins. These studies collectively demonstrate the antimicrobial properties of tannins and their potential applications in packaging materials for inhibiting the growth of microorganisms and extending the shelf life of food and beverage products.

### 5.3. Color Stabilization Properties

Tannins are compounds that can form  $\pi$ -stacking interactions with aromatic compounds. They can potentially be used as “co-pigments” to improve the storage and heat stability of anthocyanins. Tannins help preserve the color intensity in red wine and food and beverages made from fruits and berries by preventing water from adding to the flavylum ion (Moccia et al. 2020a; Trouillas et al. 2016). Then, the addition of tannin and yeast extracts could significantly improve the content of total anthocyanin and C3R (cyanidin-3-rutinoside) in mulberry wine and thus positively influence their stability (You et al. 2018). Another study investigated the influence of oenological tannins on the effectiveness of color stabilization in red wines during aging. Indeed, hydrolyzable tannins and, more specifically, gallotannins are the most efficient compounds for stabilizing the color of red wines during aging by co-pigmentation. The mechanism of pigment formation or co-pigmentation differs concerning the botanical origins of oenological tannins since they present different chemical structures. In this way, condensed tannins can combine directly or indirectly with anthocyanins, while hydrolyzable tannins cannot participate in condensation reactions with anthocyanins. However, hydrolyzable tannins have the potential to modulate oxidation-reduction phenomena in wines, thereby protecting wine anthocyanins from oxidation and participating in co-pigmentation reactions (Vignault et al. 2022).

## 6. Future Perspective

Using tannins as active packaging materials has various potential developments and predictions in the future. Here are some perspectives on how the use of tannins as active packaging may develop:

- Packaging material innovations: In the future, we can expect innovations in packaging materials that combine tannins with other materials, such as bioplastics or nanomaterials, to create stronger, lighter, and environmentally friendly packaging. This can help reduce plastic waste and improve sustainability in the packaging industry. Besides that, this versatility allows for the development of innovative packaging designs with tailored properties suitable for specific products (Ferri et al. 2023; Nile et al. 2020).
- Better product maintenance: Further research could lead to the development of more specific tannins, which can be tailored to maintain the quality of food and beverage products in better condition. This will help reduce the wastage of food and beverages due to loss of quality (Versino et al. 2023)
- Wider pharmaceutical applications: In the pharmaceutical field, tannins can be used in active packaging to protect drugs and vaccines from oxidative and microbial damage. This can increase the shelf life of important pharmaceutical products (Zongo et al. 2021).
- Research on bioactive properties: More research may be conducted to understand better and utilize tannins' bioactive properties. This could include research into the potential use of tannins in preventing disease or supporting human health through packaged foods and beverages (Oluwole et al. 2022).

- Circular-based active packaging: Tannins can be extracted from various plant sources, including by-products of the agriculture and forestry industries. Using tannin in packaging could provide an additional economic incentive for utilizing these waste materials, contributing to a more sustainable production cycle (Singh et al. 2022).

The utilization of tannins in active packaging continues to grow with further research and innovation in this field. This can enhance the quality and shelf-life of products, reduce the adverse environmental effects of the packaging sector, and enhance human health by providing safer and higher-quality food and beverages.

## 7. Conclusions

Tannins are secondary metabolites present in plants that plants synthesize. Its availability in plants is quite diverse, but some are scattered in almost all plants. Tannins can be found in several sources of tree bark, seeds, leaves, fruit, roots, and rhizomes. Tannins are categorized into two types, namely condensed and hydrolyzable tannins. Tannins have phenolic groups, which can cause tannins to have functional properties, namely as antioxidants, antibacterial, and anti-inflammatories. The antioxidant and antimicrobial properties in tannins can be used as active ingredients in food preservatives, modified into materials for food packaging, and compounds that can be utilized in the food industry. Tannins have several other advanced applications, such as the functional food, pharmaceutical, and wood industries. Tannins can be used in polymer material so that their physical properties are close to plastic ones. They can also be coupled with the bioactive properties of tannins to be used as bioactive packaging that is easily degraded. One example is the packaging of nanocellulose-tannin base materials. However, incorporating tannins into polymers requires an appropriate method to produce polymer-tannin composites with good physical and functional properties compatibility. Several methods of combining tannins with other polymers are the one-step reactive extrusion method, the layer-by-layer method, the dynamic vulcanization method, the acetylation method, and the in-situ extrusion method. This literature review will help overcome or at least set research guidelines to overcome issues in the continued application of tannins in polymers, especially in developing tannin-based bioactive packaging.

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