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# REVIEW OF TANNINS CURRENTLY USED IN THE LEATHER INDUSTRY. PART 1: HYDROLYSABLE TANNINS

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Tannins are water-soluble phenolic substances obtained from plants by various extraction methods. Tannins can be found in almost every plant. However, tannin content varies from plant to plant. Even different parts of the same plant may contain different amounts of tannin. Tannins can be found in various parts of plants such as leaves, roots, fruits, wood, galls and bark. Tannins have many different kinds of uses in the food, cosmetics, painting, pharmaceutical, wood adhesive, and other industries due to their antibacterial, antioxidant, and other qualities. On the other hand, they have long been used in leather production due to their ability to chemically react with the leather protein collagen. Dicotyledonous plants are generally used in leather tanning due to their rich tannin content. According to a common classification, tannins are divided into three classes: Hydrolyzable, Condensed and Complex tannins. Of these, hydrolyzable tannins are hydrolyzed in the presence of various chemicals (dilute bases, dilute acids and enzymes) and are divided into 2 subclasses as gallotannins and ellagic tannins. This review includes the general chemical structures of hydrolyzable tannins. In addition, hydrolyzable plant tannins used in leather production (sumac, chestnut, myrobalan, valonia and tara) and the properties they impart to the leather are explained.

Keywords: leather, tannins, hydrolysable tannins

## INTRODUCTION

Tannins are phenolic compounds with the molecular formula $C_6H_5OH$ that are present in many different parts of higher plants, including the bark, leaf, stem, fruit, root, and wood (Redwood, 2020). Tannins participate in the defense mechanisms of plants, protecting them from insects and microorganisms. Since tannins are typically found in plant growth regions like secondary phloem, xylem, and the layer in root tissue between the cortex and epidermis, it is also believed that they can help regulate the growth of these tissues (Pizzi *et al*., 2024).

Tannins can chemically interact with proteins to form complexes (Pizzi, 2019). They are derived from plants using a variety of extraction techniques or are directly ground into powder for have been used for centuries in the tanning and retanning of leather. Besides, tannins can be used as dyes because they can give color to the leather. In addition, they can eliminate free formaldehyde that occurs on the leather due to release by resins and/or other chemicals (Bayramoğlu, 2013; Bayramoğlu *et al*., 2008; Çolak *et al*., 2004; Mirzamuratova *et al*., 2024a). On the other hand, tannins are a group of secondary metabolite polyphenols known as natural antioxidants and exhibit UV protective properties and so they can be inhibitors of lipid peroxidation and prevent Cr(III) from converting to Cr(VI) (Mirzamuratova *et al*., 2024b; Ismayati *et al*., 2024).

These properties of tannins show that they are highly functional materials and make them industrially attractive. In this study, the general chemical structures of hydrolysable tannins and hydrolysable vegetable tannins commonly used in the leather industry are mentioned.

### Chemical Structure of Tannins

According to a generally accepted classification, tannins are divided into three parts as "hydrolysable", "condensed" and "complex". Hydrolysable tannins are also classified as two


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subunits as "gallotannins" and "ellagic tannins". Gallotannins are polyesters of glucose and gallic acid that are commonly found in nature and when they are hydrolyzed, gallic acid is released (Molnar *et al*., 2024; Okuda & Ito, 2011; Porter, 1992; Hemingway & Karchesy, 2012; Seigler, 1998). Ellagic tannins are characterized by a glucose center esterified with at least one unit of hexahydroxydiphenyl acid, formed by the oxidative bonding of two units of gallic acid (Mavlyanov *et al*., 2001; Auad *et al*., 2020; Schofield *et al*., 2001; Porter, 1989; Mueller, 2001; Ekambaram *et al*., 2016; Romani *et al*., 2006).

## Hydrolysable Tannins

Hydrolyzable tannins are a class of compounds with polyols as core and phenolic carboxylic acids connected by ester bonds, $C_6$–$C_1$ type polyphenols. Therefore, they are susceptible to hydrolysis of ester bonds in the presence of dilute bases, dilute acids or enzymes (Guo *et al*., 2024). The molecular weights of hydrolyzable tannins range from simple glycogaline (MW of 332 Da) to pentameric ellagitannins, and their MWs are above 5000 Daltons (Da) (Molnar *et al*., 2024).

### *Gallotannins*

It is composed of a gallic acid and a polyol (in most cases D-glucose) whose hydroxyl functions can be replaced by one or more gallic groups. Gallotannins are found in many plants as TGG (2,3,4,6-tetra-O-galloyl-D glucopyranose) and β-PGG (1,2,3,4,6-penta-O-galloyl-β-D glucopyranose). They serve as basic compounds for the biosynthesis of hydrolyzable phenolic compounds (Ben Aziz *et al*., 2024).

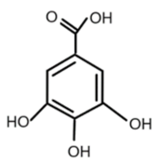


Figure 1. Gallic acid molecule

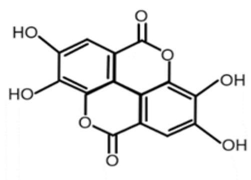


Figure 2. Ellagic acid molecule

### *Ellagic Tannins*

Ellagic tannins are biosynthetic products of gallotannins. The most important acyl group, Hexahydroxydiphenic (HHDP) group, is produced by oxidative coupling between two galloyl groups. Some other ellagic tannin acyl groups are also produced by oxidative coupling of galloyl and HHDP groups (Liu, 2024).

Because there are so many phenolic hydroxyl groups in close proximity to one another, they are very astringent; in this sense, gallotannins are more astringent than ellagitannins. The acidity of the carboxylic acid groups in the hydrolysable tannins-for example, the pH values of valonia, sumac and chestnut solutions are 3.2, 3.7-4.2, and 2.6-2.8, respectively-contributes to their reactivity. As a result, the less acidic tannins act as self-buffers (Covington, 2009).

Leathers tanned using these tannins shrink at a temperature between 75 and 80 °C. Given that this is a defining characteristic of hydrolyzed tannin (Covington, 2009).

## Hydrolyzable Tannins Widely Used in Leather Production

The interaction between tannins and proteins is widely used in leather production. Tannic acid (TA), which is abundant in hydroxyl groups that can oxidize to quinones, improves film properties by establishing covalent bonds (C=N or C-N) with amino acid residues of proteins (Choi & Kim, 2020). In contrast, under acidic or neutral pH conditions, reversible non-covalent bonds can be formed between proteins and polyphenols, including hydrogen bonds (Figure 3), hydrophobic interactions, and electrostatic interactions. In particular, oxidized TA has been reported to improve the mechanical properties of collagen films (Zhang *et al*., 2010; Zhao *et al*., 2024).

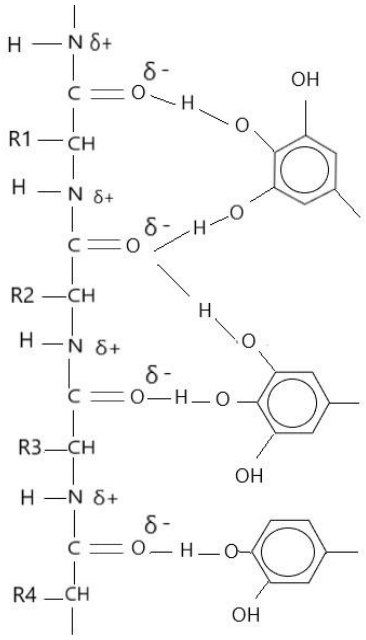


Figure 3. H bonds between the polypeptide chain of collagen and polyphenols (Covington, 2009)

Tannins form hydrogen and covalent bonds with functional groups (e.g. COOH, $–NH_2$) of the leather protein collagen. However, these bonds only occur if the tannin molecules are large enough to bind to neighboring collagen chains and contain enough phenol to form cross-links in several places. The vegetable tanning process consists of two stages: penetration and fixation. Penetration refers to the diffusion of tannins into the leather, while fixation ensures that the penetrated tannins bind to the collagen, which forms the stable material. In this process, different components such as temperature, pH, mechanical effects and particle size come into play to obtain the desired products (Tasnim *et al*., 2024; Mia *et al*., 2024). Sumac, chestnut, myrobalan, valonia and tara are hydrolysable tannins that are generally used commercially in terms of tanning efficiency, quality, performance and extraction efficiency.

Table 1. Hydrolysable tannins commonly used in leather production and their properties (Redwood, 2020)

| Plant | Source of tannin | Tannin content (%) | Non-tans content (%) |
|---|---|---|---|
| Sumac (*Rhus coriaria /Rhus cotinus*) | Leaves | 22-35 | 14-15 |
| Chestnut (*Castanea sativa*) | Wood | 5-15 | 16-17 |
| Myrobalan (*Terminalia chebula*) | Nuts | 25-48 | 14-17 |
| Valonia (*Quercus Aegilops*) | Fruit | 4-10 | 1-2 |
| Tara (*Caesalpinia spinosa*) | Pods | 30-35 | 1-2 |

*Sumac*

Sumac tannins are produced from the leaves of shrubs belonging to the *Rhus coriaria* genus, which is native to Southern Europe and the Mediterranean (Türkiye, KKTC (Turkish Republic of Northern Cyprus), Greece, Bulgaria, Croatia, Montenegro, Italy). Sumac tannins belong to the gallotannin family of hydrolyzed tannins. They are highly sensitive to heat compared to other tanning agents and break down into gallic acid when exposed to heat. Sumac leaves contain approximately 30% tannin. They also contain gallic acid, glucose, chlorophyll and inorganic salts. Leather tanned with sumac tannin is light and soft. It has a pleasant touch and a smooth texture. This type of leather can be dyed with both anionic and basic dyes. It has high light, oxidation and sweat resistance (Shabbir, 2012; Falcão & Araújo, 2011; Plavan *et al*., 2010).

*Chestnut*

*Castanea sativa* belongs to the *Fagaceae* family and is among the most common chestnut species (De Vasconcelos *et al*., 2007). The main functional units in the wood, bark and fruit of the chestnut tree (*Castanea sativa L*.) are hydrolyzed tannins such as gallic acid and ellagic acid (Buyse *et al*., 2021). In addition to these main units, subunits such as castalin, vescalin, castalagin, vescalagin, kurigalin, 5-O-galloylhamamelose, (3′,5′ dimethoxy-4′-hydroxyphenol)-1-O-β-D-(6-O-galloyl) glucose, chestanin and acutissimin A have been recorded in chestnut extract. Chestnut extract is commercially used in animal feed, leather production and the food industry (wine and alcoholic beverages) (Comandini *et al*., 2014).

The standard extract has a pH of 3.5; inorganic salts and hydroxides are added to create a "sweetened" extract with a pH of 4.5. Softer leather is produced by the less astringent nature of the sweetened extract. It can be used alone or in conjunction with syntans to retan chrome leather and tanning sole leather. These combinations result in leather that is lighter in color and has superior tanning qualities (Krisper *et al*., 1992). Sweetened chestnut penetrates the leather better. Leathers tanned with sweetened chestnut are more yellow than those tanned with normal extract. On the other hand, leathers produced with normal chestnut extract are firmer, fuller, harder, and more resistant to water and abrasion (Yahia *et al*., 2019).

*Myrobalan*

Myrobalan is the unripe fruit of the *Terminalia chebula* tree. It is in the hydrolyzed tannins class. Its structure contains compounds that are esters of glucose such as phenol carboxylic acid and 1,2,3,4,6-pentagalloyl glucose. It contains around 35-40% tannin. Its tanning effect is light, it is generally used as an additive in vegetable tanning. Leathers produced with Myrobalan are light in color (Sivakumar *et al*., 2018).

*Valonia*

Oak, which belongs to the *Fagaceae* family, is common in temperate climates in some parts of Europe, Asia and North Africa. Its fruits are called acorns and grow in groups of 2-5 on a stem. Known as a high-calorie (339 kcal/100 g) nutrient, acorn seeds are rich in vitamin C, magnesium and calcium. Other active substances found in acorns are quercitanic acid, ellagic and gallic acid, tannin, quercin, fluoroglycine, pectic substances, resins, calcium oxalate, pentadigalloylglucose, cyclogallifaric acid and carbohydrates. These substances provide astringent, antiseptic, anti-inflammatory and antioxidant properties to acorns (Simion *et al*., 2023).

Oak trees of the *Quercus* species, which have different varieties, are generally used in furniture making and construction materials. Tannins can be obtained from wood, bark, gall and acorn. Depending on the species, there is 50-70% tannin in galls, 10-20% in shells, and approximately 10% in acorns (Baytop 1999; Bayramoğlu, 2012). The tanning material obtained from acorns is called 'Valex'. The acorns of the *Quercus aegilops* tree, also known as the Turkish oak, which grows abundantly in Türkiye, Greece and neighboring countries, are usually collected in August and dried in domes. It is widely used in leather production in Austria, Germany and France, allowing faster, harder, tighter, heavier and waterproof leather production. The main components of valex are ellagitannins, which are castalagin, vescalagin and pentagalloylglucose. This material is often used mixed with ground oak bark in the production of shoe soles to increase quality and durability (Falcão & Araújo, 2018).

*Tara*

Tara *(Caesalpinia spinosa (Molina) Kuntze*), which is widespread in South America and Africa, is found in the form of a small legume tree or thorny shrub. The world's largest producer and exporter (about 80% of production) is Peru, and production takes place in different semi-arid regions of this country. It contains gallotannic tannins, and is grown for leather production and food use. Pre-Inca civilizations, whose origins are based on the Andean Region, used the fruits of the tara tree to produce dyes for textiles and ceramics, tanning agents for leather, and medicine. Due to the light color and light resistance of the leathers tanned with tara, it is widely used especially in automobile upholstery (Ibieta & Penarrieta, 2021; Castel *et al*., 2013).

What distinguishes tara tannin from other commercial tannins is that it is not an extract but a finely ground powder obtained from tara pods. Therefore, it has a high amount of insoluble matter (Gaidău *et al*., 2014). The tannin content varies between 30-35%. Tara gives fullness and softness to the leather. The resistance of the leathers tanned with tara to tear strength is higher than those obtained with other vegetable tannins (Aravindhan *et al*., 2015).

## CONCLUSION

Various plant tannins are used in leather production. These tannins are divided into 3 classes: hydrolyzed, condensed and complex. Hydrolyzed tannins are also divided into 2 subclasses: gallotannin and ellagic tannins. They are susceptible to hydrolysis of ester bonds in the presence of dilute bases, dilute acids or enzymes. They have the ability to react with proteins. Thus, they have been used for leather tanning for many years. For this purpose, the most commonly used hydrolyzable tannins in the market are sumac, chestnut, myrobalan and tara. Tannin from sumac leaves gives the leather smoothness. In addition to chestnut extract, there are also chemically treated chestnuts called sweetened chestnuts on the market. The leather is better penetrated by sweetened chestnut. When using sweetened chestnut extract instead of regular extract, leathers get a deeper yellow color. However, leather made using regular chestnut extract is harder, fuller, firmer, and more resilient to abrasion and water. Myrobalan is often used in combination with other tanning agents. Valonia makes it possible to produce leather that is waterproof, heavier, tighter, faster, and harder. Tara is applied directly, without the need for other tanning agents, by powdering the pods. Leathers tanned with tara are light colored and have high light fastness. Therefore, tara is popular in the creation of light-colored leather and also suitable for pastel shades. In general, when vegetable tanning agents are used alone, desired quality leathers cannot be obtained. The reasons for this are; they give color to the leather, have a low shrinkage temperature and are hydrophilic. Therefore, they are open to research in terms of improving these properties.

In recent years, sustainable production has gained importance in the leather industry. Vegetable tanning agents are natural products and are very easy to break down in nature.

Leather processed with vegetable tanning agents has a special interest for ecological leather production. In addition, plant tannin production provides employment opportunities for many people. Tannins can be used in different places and for different purposes in leather processing. The advantages of vegetable tanning agents for leather can also be presented as a separate article.

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