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Hardwood Tannin: Sources, Utilizations, and Prospects

Atanu Kumar Das, Md. Nazrul Islam, Md. Omar Faruk, Md. Ashaduzzaman, Rudi Dungani, Enih Rosamah, Sri Hartati and Alfi Rumidatul

Abstract

Tannins are found in widely distributed species of plants, and it protects plant from predators and pests. There are three major groups of tannins, that is, hydrolyzable, complex, and proanthocyanidins. Tannins are being used as a significant element for the tanning of animal hides in the leather production industry from the beginning of tannin industry. Then, these have been used for mineral absorption and protein precipitation purposes since the 1960s. Tannins are used for iron gall ink production and wood-based industry as adhesive and anticorrosive, recovering uranium from seawater and removing mercury and methylmercury from solution. In addition, tannins are considered as bioactive compound in nutrition science, and their possible effects on health are to be identified. This chapter outlines the structural and biological properties of hardwood tannins to indicate the positive utilization of them. It also describes the contemporary information on tannins.

Keywords: tannin, structural properties, biological properties, utilization

1. Tannin and its classification

Plants are protected from herbivores and diseases due to the accumulation of a wide range of “secondary” compounds, together with alkaloids, terpenes, and phenolics. Phenolic metabolism helps to produce a wide variety of compounds ranging from the familiar anthocyanidins (flower pigments) to lignin (complex phenolics of the plant cell wall). Generally, the chemical and biological criteria of phenolic compounds are called as tannins which are different from other plant secondary phenolics. The word “tannin” originates from the ancient Celtic word for oak [1].

Seguin has introduced the term “tannin” to explain the ability to convert hide or skin into leather by organic material extracted from certain plant tissues in 1796 [1]. Tannin is a chemical component obtained from plant, and it has been used for the benefit of human being from the beginning of its development. To get the idea about its complete nature, a lot of research work is needed till today, and that is why it is considered the “dark continents” of science. Tannin has the ability to make proteins and other polymers like pectin [2].

Tannins are the main polyphenolic secondary metabolites distributed widely in the range of 5–10% of dry plant material in vascular plants. They are found mainly in the bark, stems, seeds, roots, buds, and leaves [3]. Tannins are either galloyl

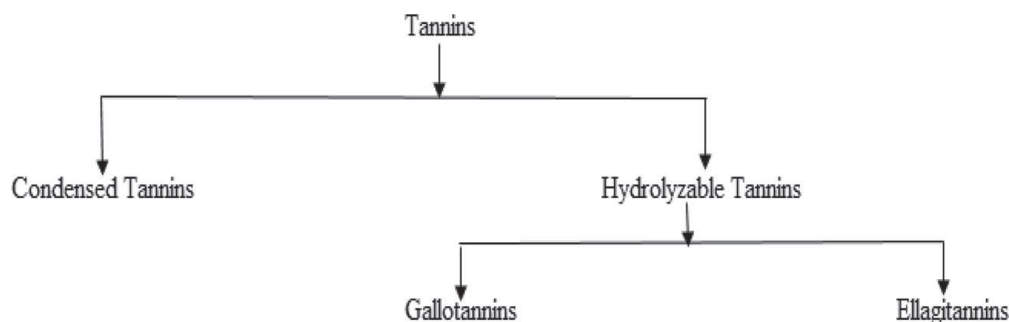


Figure 1.
Classification of tannins.

esters, or they are oligomeric and polymeric proanthocyanidins. They protect trees from fungi, pathogens, insects, and herbivorous animals [4].

Tannins in softwood and hardwood are different as softwood and hardwood dust particles can be separated based on their tannin content [5].

The term “tannin” is difficult to define concisely as it is found in plant chemistry. Generally, tannin contains precise physical and chemical components, which help to convert hide or skin of animal into impermeable non-rotting leather. Broadly, the definition of tannin covers a whole mass of components which give overall phenolic reactions [2].

Bate-Smith has defined tannins in this way: “tannins are water soluble phenolic compounds with molecular weight of 500 and 3000, capable of precipitating gelatin, alkaloids and other proteins and provide the typical phenolic reactions” [1, 6]. Tannins having a high molecular weight of 20,000 have shown complex with some specific polysaccharides [1, 6]. Scientists have also used the term “polyphenol” in place of “tannin” to emphasize the diversity of phenol groups which characterize these compounds [7, 8].

Natural tannins are generally categorized as either condensed or hydrolyzable tannins (**Figure 1**). Hydrolyzable tannins are subdivided into gallotannins and ellagitannins [4].

Condensed tannins have a wide range of molecular weight from 500 to over 20,000 [9]. Condensed tannins are composed of flavanols or polymers of flavan-3-ols (catechins) and/or flavan 3:4-diols (leucoanthocyanidins) [10]. Phlobaphene, a water insoluble product, is possible to be produced by polymerization of condensed tannins. Condensed tannins have the ability to react with aldehydes to generate polymeric materials [9]. The hydrolyzable tannins contain glucose or other polyhydric alcohols esterified with gallic acid (gallotannins) or hexahydroxydiphenic acid (ellagitannins) [11]. Acid hydrolysis of hydrolyzable tannins helps to get gallic acid, ellagic acid, or other similar species [7]. The most important condensed tanning materials are wattle or “mimosa,” quebracho, mangrove, and hemlock, whereas the hydrolyzable tanning materials are chestnut wood (*Castanea sativa* and *C. dentata*) and dry myrobalan fruits (*Terminalia chebula*) [10].

2. Hardwood tannin

2.1 Condensed tannin

Condensed tannins are polymeric flavonoids, and most of them are based on the flavan-3-ols (**Figure 2**) (–)-epicatechin and (+)-catechin [1].

The best categorized condensed tannins are linked by C8 of the terminal unit and C4 of the extender. The four common modes of coupling are called B-1, B-2,

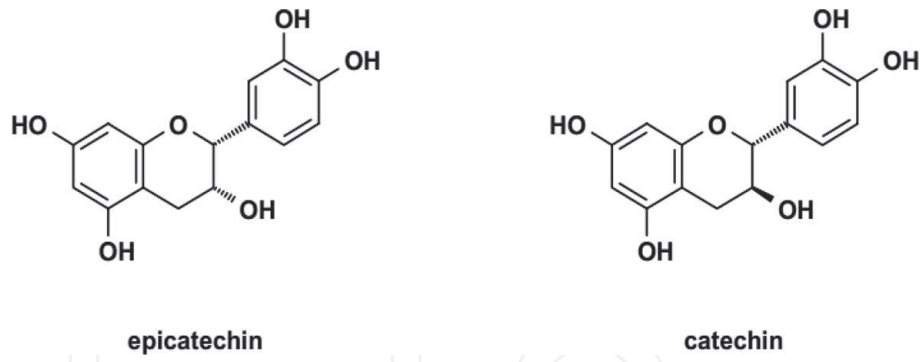


Figure 2.
Flavan-3-ols [1].

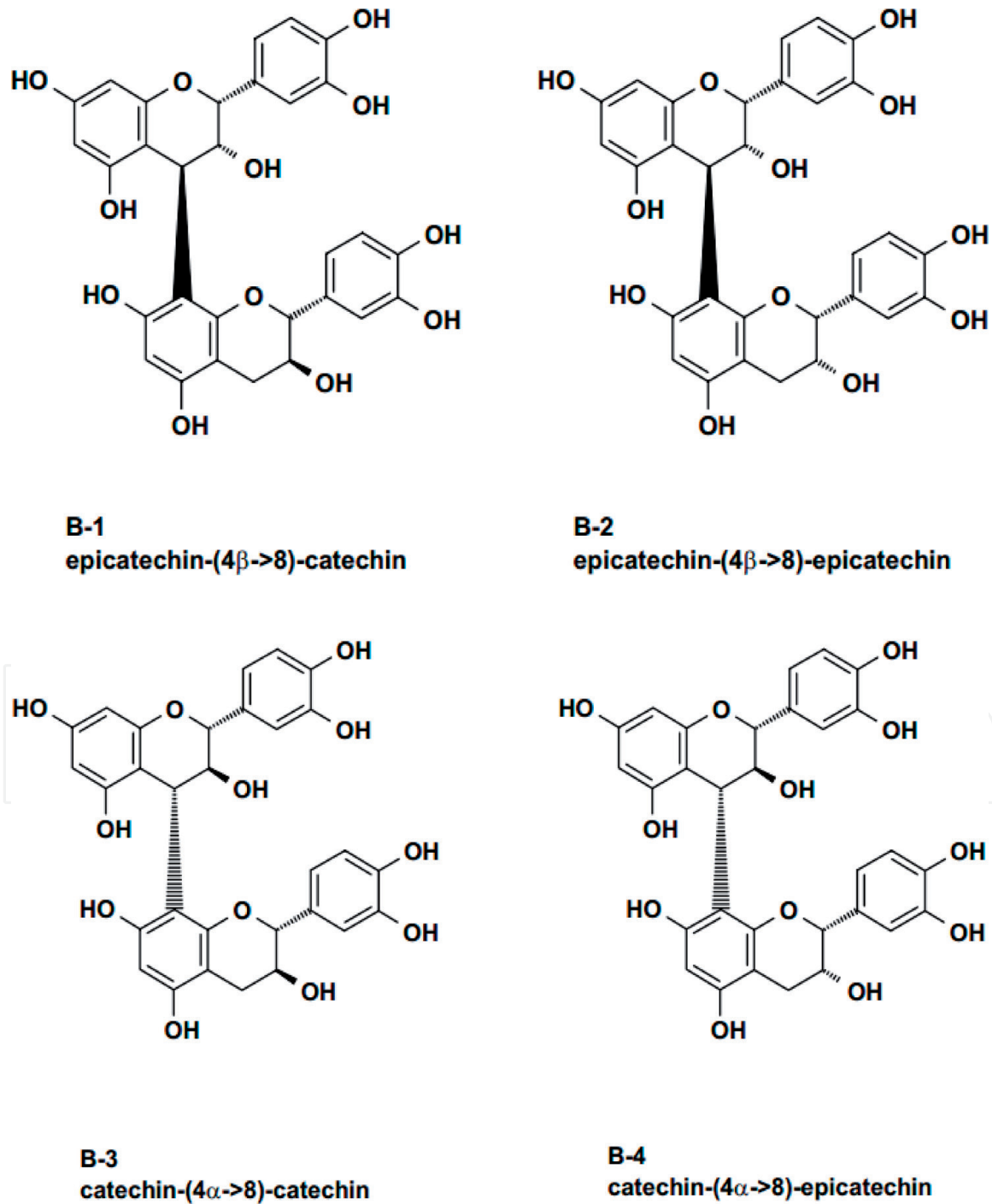


Figure 3.
Common modes of coupling of condensed tannins [1].

B-3, and B-4 (**Figure 3**). Besides these dimers, related dimers which are linked by C6 of the terminal unit and C4 of the extender have been separated [1].

Proanthocyanidin is becoming more popular to describe flavonoid-based polyphenolics instead of condensed tannins. Anthocyanidin pigments are derived from proanthocyanidins by oxidative cleavage in hot alcohols (**Figure 4**) [1].

Unmodified terminal units are produced by the acid butanol reaction, and the extender units produce the colored anthocyanidins. Procyanidins are produced by catechin- and epicatechin-based polymers. Delphinidin is the yield of gallo catechin- and epigallocatechin-based polymers, whereas pelargonidin is the product of the rare mono-substituted flavan-3-ol-based polymers.

There is an available branch in 5-deoxy-flavan-3-ol polymer-condensed tannins due to having the reactivity of the 5-deoxy A ring, and these are an important group of condensed tannins. In the case of quebracho and acacia tannin preparations, profisetinidins and prorobinetinidins are observed as the major tannins. Acid butanol reaction helps to produce the 5-deoxy anthocyanidins fisetinidin and robinetinidin (**Figure 5**) [1].

Tannins are broadly available in all over the plant kingdom, while the condensed tannins are distributed in the Archichlamydeae division of the

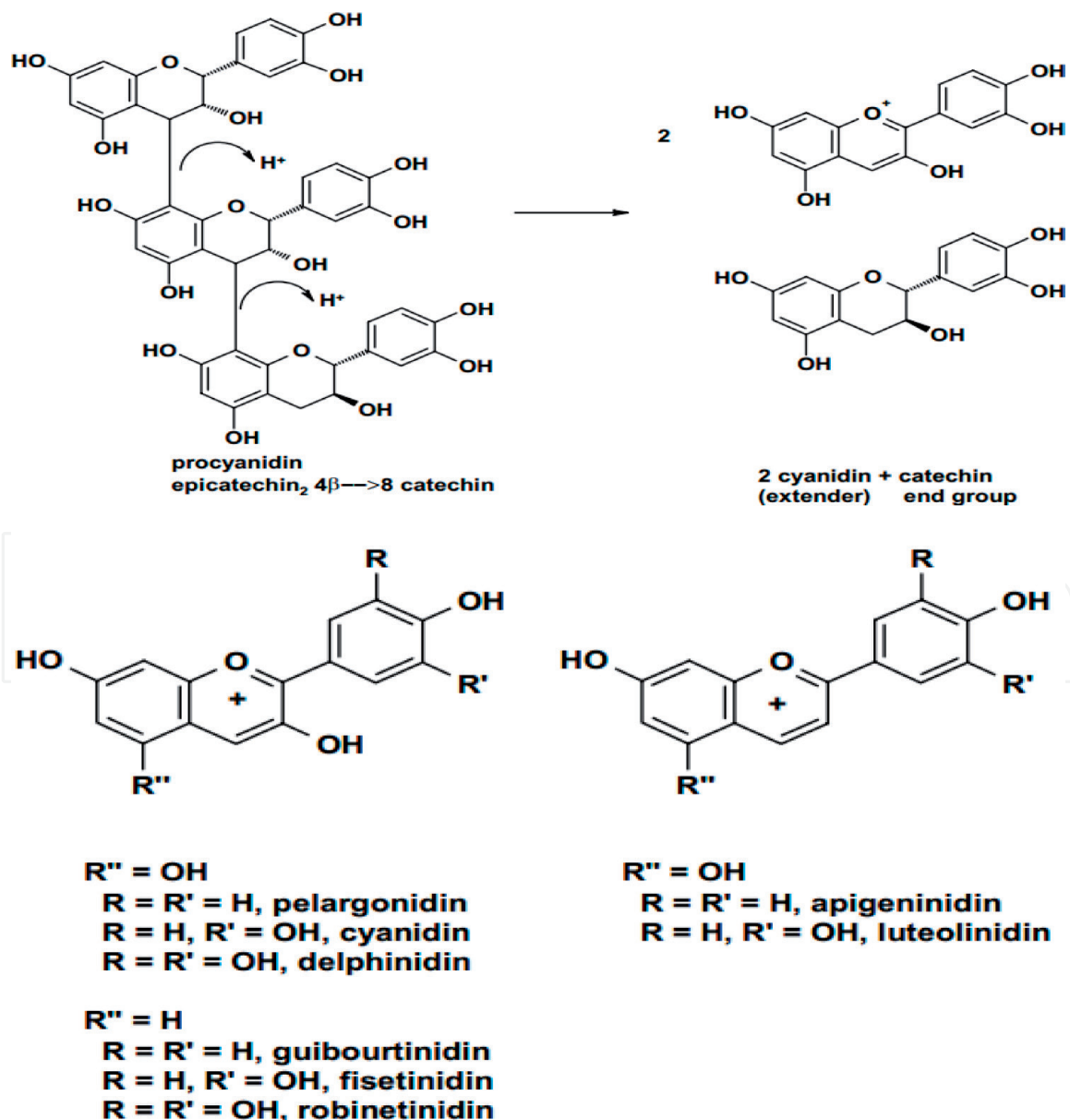


Figure 4.
Proanthocyanidin and its derivative [1].

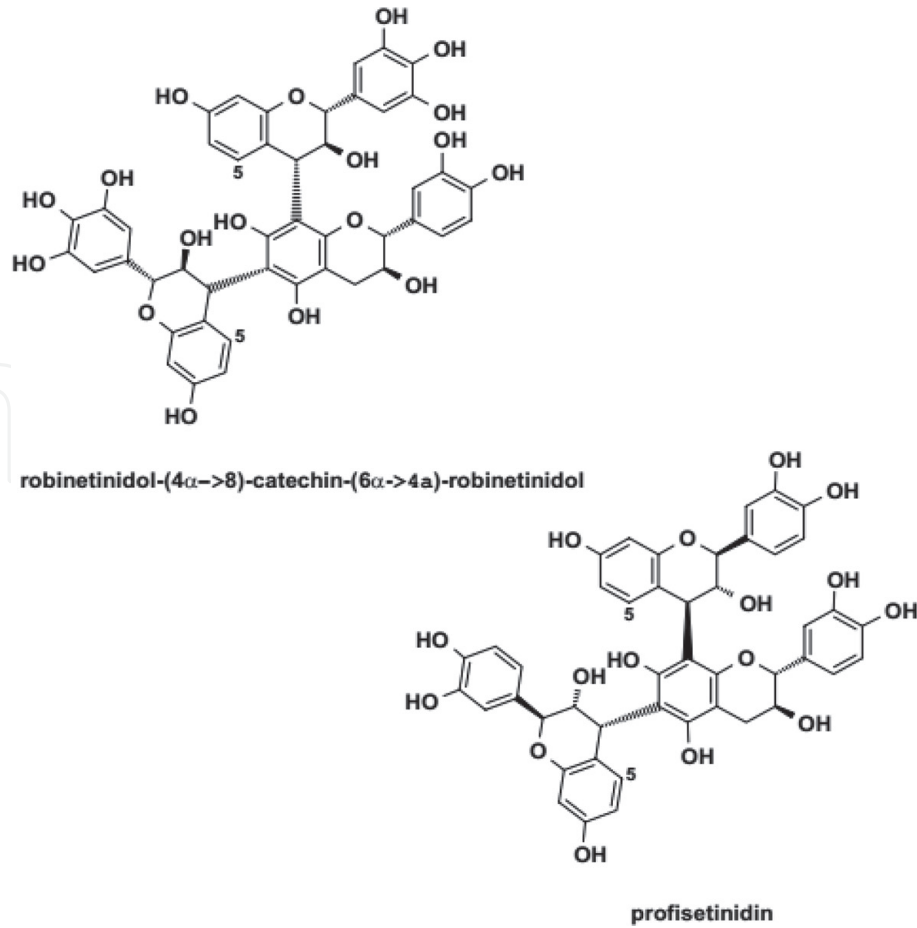


Figure 5.
 Extracted tannins by acid butanol reaction [1].

Family	Genus
Casuarinaceae	<i>Casuarina</i>
Salicaceae	<i>Salix</i>
Betulaceae	<i>Betula</i>
Fagaceae	<i>Castanea, Lithocarpus, Quercus</i>
Lauraceae	<i>Persea</i>
Rosaceae	<i>Prunus</i>
Leguminosae	<i>Acacia, Cassia, Robinia</i>
Meliaceae	<i>Carapa, Xylocarpus</i>
Euphorbiaceae	<i>Excoecaria</i>
Anacardiaceae	<i>Astronium, Rhus, Schinopsis</i>
Bombacaceae	<i>Camptostemon</i>
Sterculiaceae	<i>Heritiera</i>
Dipterocarpaceae	<i>Hopea</i>
Lythraceae	<i>Pemphis</i>
Sonneratiaceae	<i>Sonneratia</i>
Rhizophoraceae	<i>Bruguiera, Carallia, Ceriops, Candelaria, Rhizophora</i>
Combretaceae	<i>Laguncularia</i>
Myrtaceae	<i>Eucalyptus, Tristania</i>
Verbenaceae	<i>Avicennia</i>

Table 1.
 Condensed tannins in hardwood.

Dicotyledonae. Scientists examined condensed tannins by paper chromatography in their previous studies [12–16]. In Australia, the attractive source of condensed tannins are extracts from barks of *Bruguiera* and *Rhizophora* species (mangrove species) and *Eucalyptus astringens* (brown mallet) and the heartwood and bark of *E. wandoo* and *E. accedens* (wandoos). **Table 1** shows the distribution of condensed tannins in hardwood.

2.1.1 Utilization of condensed tannin

2.1.1.1 Tanning industry

In general, condensed tannin material is the major component of commercial tanning chemical, which is used for tanning leather in tannery. The major contributors for the world's supply of condensed tannin are extract of mangrove and wattle and quebracho used in heavy leather manufacture. Condensed tannin materials are selected based on their tanning quality for heavy leather production. Condensed tannins have high resistance power to detanning [17]. Quebracho and wattle tannins are more resistant to concentrated urea solution than to hydrolyzable tannins [18].

Though tannin materials are dissimilar based on pH, salt content, and natural conditions of acid content, it can be controlled maintaining proper conditions [19, 20]. pH in tan liquors is relatively high due to the absence of phenolic in tan liquors. Mangrove extracts contain sodium chloride, whereas sulfited quebracho extracts possess sodium sulfate, and the salt content contributes to the salt material in tannin materials [21].

2.1.1.2 Preservative for fishing net

Preservation of fishing nets by various condensed tannin materials is popular in Indo-Pacific countries. It is used to prevent cellulose degradation by bacterial and fungal cellulases. Fishing nets are submerged in hot tannin solution, and this method is repeated for several times. Tannin-impregnated nets are further treated with either hot ammoniacal copper sulfate or dichromate solution for some cases [22, 23]. Condensed tannin materials are extracted from Burma cutch from *Acacia catechu* heartwood, Malayan (mangrove) cutch from *Carapa obovata* and *Ceriops candolleana* barks, and Borneo (mangrove) cutch from *Ceriops candolleana*, *Rhizophora candelaria*, and *R. mucronata* barks [21].

2.1.1.3 Preparation of plastics and adhesives

Condensed tannins are phenolic raw materials which react with formaldehyde; these can be used for the production of synthetic resins [21]. Nico [24] studied the suitability of quebracho-formaldehyde resins for adhesives and plastics. Molding powders are produced by pressing mixtures of accurate quebracho and wattle tannins, paraformaldehyde, and plasticizer [21]. Though these moldings have short flow period, these follow all the characteristics of phenol-formaldehyde moldings as per the specifications of British standards [25]. These molding powders show very stable and waterproof properties, and the mixture is catalyzed with either acid or alkali at temperature 250°F [25].

Various condensed tannins as plywood and particle board adhesives have been studied by many researchers [26–30]. Adhesives prepared from bark of *Tsuga heterophylla*, *Acacia mollissima*, and *Callitris calcarata* have less strength and higher water resistance properties than those obtained from commercial adhesives [31, 32]. Strong and water-resistant adhesives for plywood are prepared by mixing of paraformaldehyde, filler, and sulfited tannin solutions obtained from *Eucalyptus crebra*, *Pinus radiata*, wattle extract,

and quebracho extract [25, 33]. Adhesives prepared from Australian mangrove tannin extracts in addition with a small proportion of commercial phenol-formaldehyde, resorcinol-formaldehyde, or phenol- and resorcinol-formaldehyde comply with Australian and British standard for synthetic adhesives for plywood [21, 34].

2.1.1.4 Oil and ceramic industry

The main problem is to maintain the flow and suspension characteristics of bentonite oil well muds in the United States [21]. In oil well drilling, quebracho tannins are used widely to control the viscosity of the mud; annual consumption of quebracho extract is 30,000–40,000 tons for this purpose [35]. In the United States, it is accounted for about 40% of the total tannin consumption in 1950 [36, 37].

Scientist has reviewed the incorporation of condensed tannins into the ceramic industry. Quebracho extract enables the use of higher solid mixer in the casting of clay slip by lowering the viscosity of the clay-water mixes. Condensed tannins save the plaster molds from deterioration by increasing the tensile strength of clay casts and eliminating the silica from the mixture, with consequent saving in the deterioration of plaster molds. The suspending power of the slip is enhanced by quebracho tannins in the casting of bone China [21].

2.1.1.5 Anticorrosive of metals

Tannins protect the iron materials from sulfate-reducing bacteria by exerting a bacteriostatic action [38]. Mangrove tannin is economically viable to protect the underground iron pipes and tubes [35].

Tannate films of tannin extract is formed on iron and steel surfaces to protect it from atmospheric corrosion [25]. Actually a protective film is formed during the submersion of aluminum in tannin solutions, and it can be used for the preservation of nonferrous metal [39].

2.2 Hydrolyzable tannin

Hydrolyzable tannins are esters of a sugar with one or more polyphenolic carboxylic acids; sugar is mainly glucose, but polysaccharides or branched-chain sugars are available for some cases. The hydrolyzable tannins are generally classified based on the hydrolyzed product, and these are either gallotannins, are simple polygalloyl esters of glucose. Pentagalloyl glucose, has five identical ester linkages that involve aliphatic hydroxyl groups of the core sugar (**Figure 6**). Alkalies, acids, or enzymes like tannase and takadiastase hydrolyze the ester linkages in hydrolyzed tannins [40].

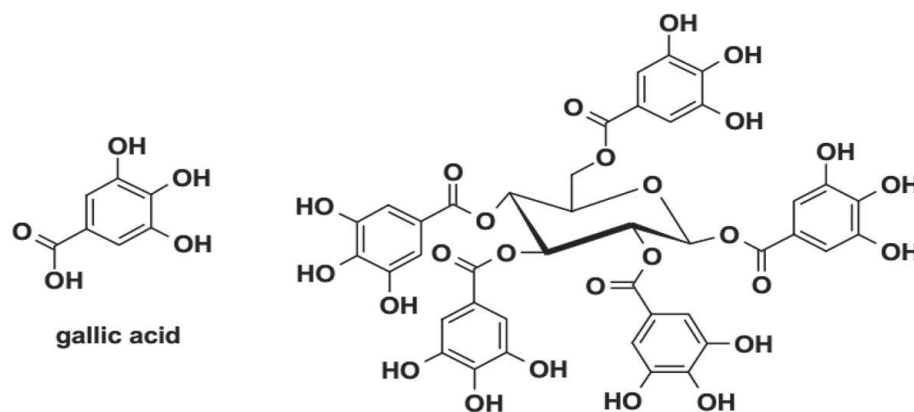


Figure 6.
Precursors of gallotannins [1].

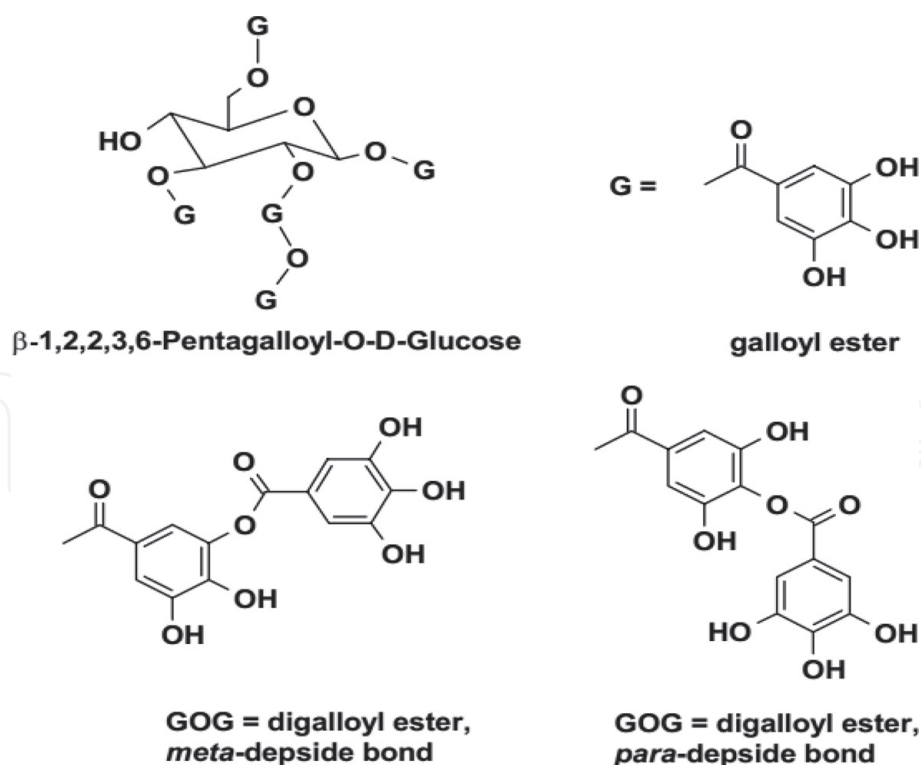


Figure 7.
Isomers of PGG [1].

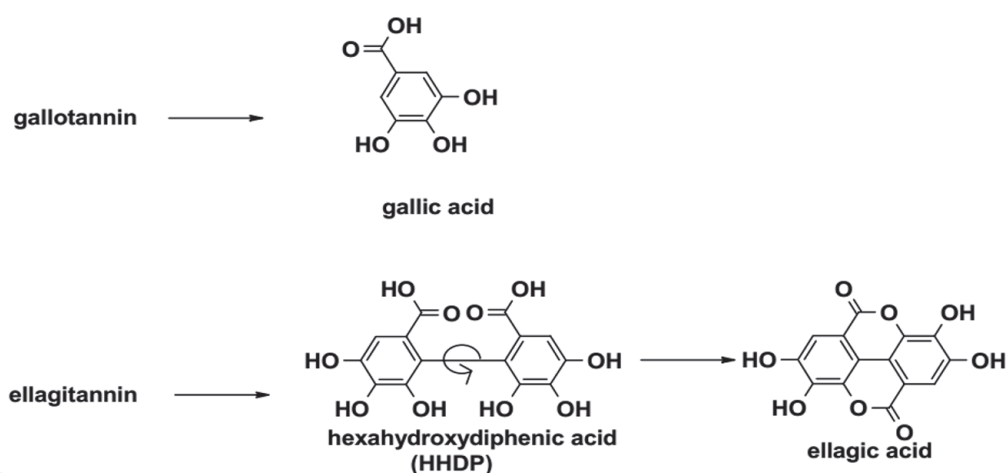


Figure 8.
Ellagitannins [1].

Gallotannins, the simplest hydrolyzable tannins, are simple polygalloyl esters of glucose. The ideal gallotannin is comprised esters of β -1,2,3,4,6-pentagalloyl-O-D-glucopyranose (pentagalloylglucose) (Figure 6). Pentagalloylglucose (PGG) has five identical ester linkages having aliphatic hydroxyl groups of the core sugar [21].

PGG has many isomers having the same molecular weights (940 g/mol) (Figure 7). Although the molecular weights are same, chemical properties and biochemical properties are different from each other due to having different structures. Therefore, their properties like susceptibility to hydrolysis, chromatographic behavior, and ability to precipitate protein are varied for different isomers [21].

Gallotannins are converted to the related ellagitannins by the oxidative coupling of galloyl groups (Figure 8). The simple ellagitannins are esters of hexahydroxydiphenic acid (HHDP). In aqueous solution, HHDP impulsively lactonizes to ellagic acid [21].

HHDP is formed by intramolecular carbon-carbon coupling C-4/C-6 (e.g., eugenin) and C-2/C-3 (e.g., casuarictin, also has C-4/C-6) having polygalloyl glucoses

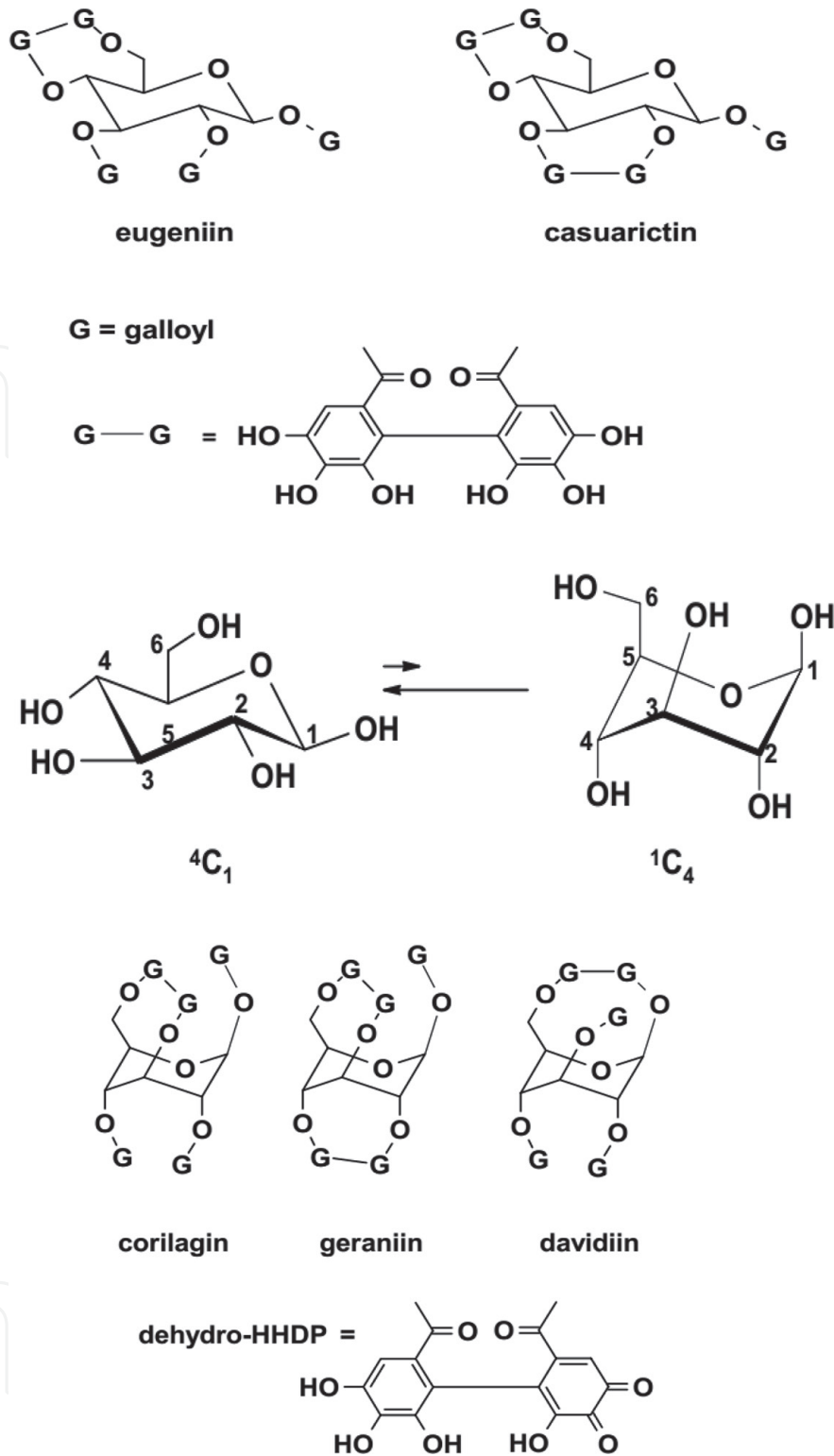


Figure 9.
 Hexahydroxydiphenic acid (HHDP) [1].

in the more stable 4C_1 conformation (**Figure 9**). On the other hand, intramolecular coupling occurs at C-3/C-6 (e.g., corilagin), C-2/C-4 (e.g., geraniin, also has C-3/C-6), or C-1/C-6 (e.g., davidiin) with polygalloyl glucose the less stable 1C_4 conformation for a few plants (**Figure 9**). In addition, geraniin is characterized by partial oxidation of the C-2/C-4 HHDP to dehydro-HHDP [21].

Some commercial hydrolyzable tannins presented in **Table 2** develop the present knowledge on the chemistry of the hydrolyzable tannins. Hydrolyzable tannins are available in the leaves and sapwood of a tree [40, 41]. Ellagitannins (or ellagic acid) are found in the leaves of species of the orders *Fagales*, *Myrtiflorae*, *Rosales*,

Tannin	Source
Chinese tannin (tannic acid)	Galls on leaves of <i>Rhus semialata</i> (Anacardiaceae)
Sumac tannin	Leaves of <i>Rhus coriaria</i> (Anacardiaceae)
Myrobalans	Fruit of <i>Terminalia chebula</i> (Combretaceae)
Turkish tannin	Galls on wood of <i>Quercus infectoria</i> (Fagaceae)
Valonea extract	Acorn cups of <i>Quercus valonea</i> or <i>Q. macrolepis</i> (Fagaceae)
Oak extract	Wood of various <i>Quercus</i> species (Fagaceae)
Chestnut extract	Wood of <i>Castanea sativa</i> and <i>C. dentata</i> (Fagaceae)
Tara extract	Pods of <i>Caesalpinia spinosa</i> (Leguminosae)
Divi-divi	Pods of <i>Caesalpinia coriaria</i> (Leguminosae)
Algarobilla extract	Pods of <i>Caesalpinia brevifolia</i> (Leguminosae)

Table 2.
Name of commercial hydrolyzable tannins and sources.

Family	Genus
Casuarinaceae	<i>Casuarina</i>
Juglandaceae	<i>Pterocarya</i>
Betulaceae	<i>Alnus</i> , <i>Carpinus</i>
Fagaceae	<i>Castanea</i> , <i>Nothofagus</i> , <i>Quercus</i>
Loranthaceae	<i>Nuytsia</i>
Nymphaeaceae	<i>Nuphar</i>
Cercidiphyllaceae	<i>Cercidiphyllum</i>
Droseraceae	<i>Drosera</i>
Saxifragaceae	<i>Francoa</i> , <i>Ribes</i>
Hamamelidaceae	<i>Liquidambar</i>
Rosaceae	Rosoideae (<i>Geum</i> , <i>Fragaria</i> , <i>Potentilla</i> , <i>Rosa</i> , <i>Rubus</i>) only
Leguminosae	<i>Pterocarpus</i> (<i>Caesalpinia</i> in lit.)
Geraniaceae	<i>Geranium</i>
Tremandraceae	<i>Tetratheca</i>
Euphorbiaceae	<i>Acalypha</i> , <i>Euphorbia</i> , <i>Ricinus</i>
Empetraceae	<i>Empetrum</i>
Coriariaceae	<i>Coriaria</i>
Cyrtillaceae	<i>Cyrtilla</i>
Corynocarpaceae	<i>Corynocarpus</i>
Aceraceae	<i>Acer monspessulanum</i> (<i>A. ginnala</i> in lit.)
Meliantaceae (Anacardiaceae in lit.)	<i>Greyia</i> , <i>Melianthus</i>
Vitaceae	<i>Vitis</i>
Elaeocarpaceae	<i>Aristotelia</i>
Theaceae	<i>Camellia</i> , <i>Cleyara</i> , <i>Gordonia</i> , <i>Thea</i>
Frankeniaceae	<i>Frankenia</i>
Tamaricaceae	<i>Tamarix</i>
Bixaceae	<i>Bixa</i>
Stachyuraceae	<i>Stachyurus</i>
Elaeagnaceae	<i>Elaeagnus</i> , <i>Hippophae</i>

Family	Genus
Lythraceae	<i>Lagerstroemia, Lythrum</i>
Punicaceae	<i>Punica</i>
Rhizophoraceae	<i>Cassipourea</i> (<i>Rhizophora</i> in lit.)
Combretaceae	<i>Combretum</i> (<i>Terminalia</i> in lit.)
Myrtaceae	<i>Agonis, Angophora, Callistemon, Eucalyptus, Eugenia, Melaleuca, Metrosideros, Myrtus, Psidium</i>
Melastomataceae	<i>Bertolonia, Heterocentron, Medinilla, Tibouchina</i>
Onagraceae	<i>Dircaea, Fuchsia, Jussieua, Lopezia, Oenothera</i>
Haloragaceae	<i>Gunnera, Haloragis, Myriophyllum</i>
Ericaceae	<i>Arbutus</i> (<i>Arctostaphylos, Vaccinium</i> , in lit.)
Diapensiaceae	<i>Galax</i>
Campanulaceae	<i>Centropogon</i> only
Compositae	<i>Tagetes</i> only
Amaryllidaceae	<i>Hypoxis</i>

Table 3.
 Name of family and genus containing ellagitannins or ellagic acid.

Sapindales, and *Geraniales* [42]. Gallotannins and ellagitannins have been detected in the tissues of many hundreds of species of dicotyledons [40]. Source of ellagitannins or ellagic acid is presented in **Table 3**.

2.2.1 Utilization of hydrolyzable tannin

2.2.1.1 Tanning industry

For leather manufacturing process, the conversion of animal hide or skin into leather is considered as the main art of tanning in leather industries. To provide leather, tannin cross-links with the collagen chains located in the hide during tanning process. Hydrolyzable tannins obtained from plant extract are widely used for tanning in leather industries [2].

2.2.1.2 Medication

Biological activities of hydrolyzable tannins confirm the beneficial effect on the health of human being, and these are used as antimutagenic, anticancer, and antioxidant. Furthermore, hydrolyzable tannins help to diminish serum cholesterol and triglycerides and suppress lipogenesis by insulin [43, 44].

2.2.1.3 Other applications

Tannic acid is used in dye industry and water treatment process for purifying water. It is also used for manufacturing ink, plastic resins, adhesives, surface coatings, gallic acid, etc. [2].

2.2.2 Prospectus of the utilization of tannins

There are several uses of tannins for the welfare of human being, and continuous research is being carried out for increasing the diversity of utilization of tannins. The potential uses of tannins are in the following.

2.2.2.1 Tanning industry

Tannins were the major tanning material for converting animal hide/skin into leather. At present, inorganic salts especially chrome salts are used to produce 70–85% leather. Replacing toxic chrome salts with less toxic and environmentally friendly tanning materials is possible by polymerization of tannins with other synthetic materials [11].

2.2.2.2 Wood composite industry

Tannins as wood adhesives have been confirmed by a number of patents. Synthetic adhesives are hazardous to environment. Therefore, tannins can be used as alternative source of adhesive in wood composite industry. Cost-effectiveness and high level of consistency should be considered for the replacement of existing harmful adhesives with tannins [11].

2.2.2.3 Water and waste water treatment plant

Tannins have a positive potentiality to be used as water treatment chemicals. For municipal water treatment, floccotant, a commercial product, has been used to eradicate suspended colloidal matter like clay and organic matter. Chelating ability of tannin is important to remove metallic ions, that is, Cr, Ni, Zn, or Cd, from waste water [11].

Acacia mearnsii cryogel obtained from the extraction of *A. mearnsii* tannin with formaldehyde in alkaline medium is used as adsorbent of heavy metal (**Figure 10**). Extraction of *A. mearnsii* tannin by epichlorohydrin in N,N-dimethylformamide medium, followed by grafting with diethylenetriamine and triethylamine, yields a cationic adsorbent. This cationic adsorbent is also used for heavy metals (**Figure 11**) [45].

A. mearnsii cryogel is also applicable for the removal of dyes. The dyes adsorption capacity by *A. mearnsii* cryogel is presented in **Figure 12**. It is noted that cationic dyes have higher adsorption efficiency than anionic dyes, like tartrazine and alizarin violet. On the other hand, cationic adsorbent has a high efficacy for many anionic dyes, even for a cationic dye such as methylene blue (**Figure 13**) [45].

Figures 14 and **15** show the various pesticides and pharmaceutical absorption capacity of cationic tannin adsorbent. Cationic compounds, like 2,4-D, clofibric acid, MCPA, acetylsalicylic acid, diclofenac, ketoprofen, naproxen, etc., are absorbed effectively, whereas cationic products, such as atrazine, amoxicillin, trimethoprim, are adsorbed ineffectively [45].

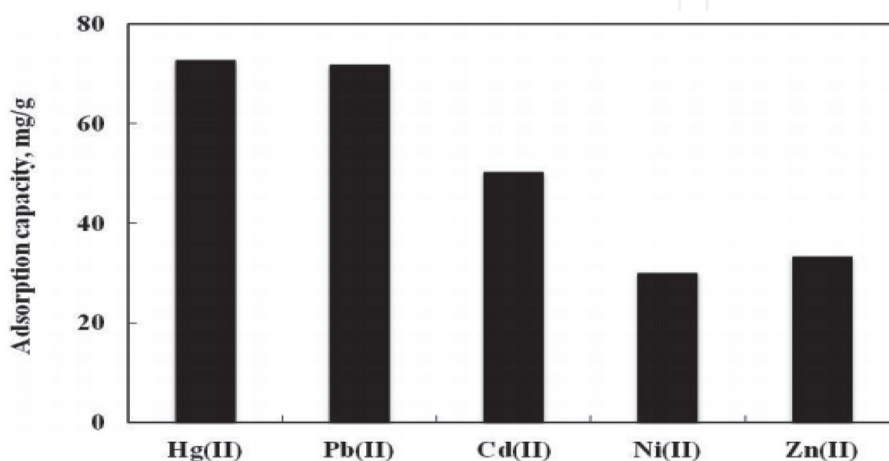


Figure 10. Comparison of the adsorption capacity of heavy metals on *Acacia mearnsii* cryogel [45].

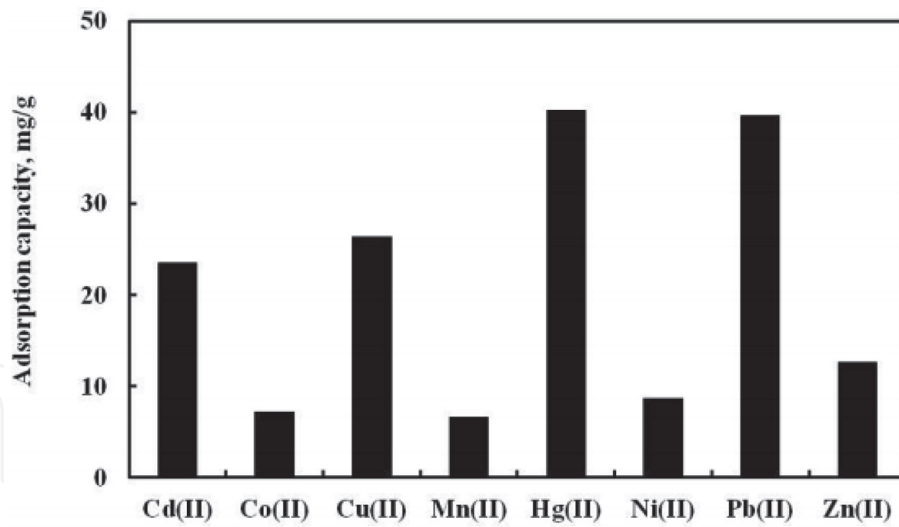


Figure 11.
 Comparison of the adsorption capacity of heavy metals on cationic tannin adsorbent [45].

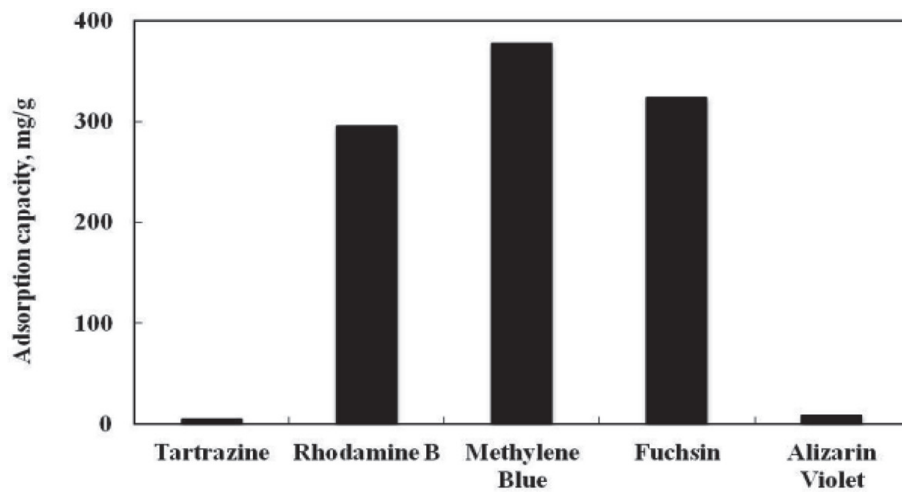


Figure 12.
 Comparison of the adsorption capacity of dyes on *Acacia mearnsii* cryogel [45].

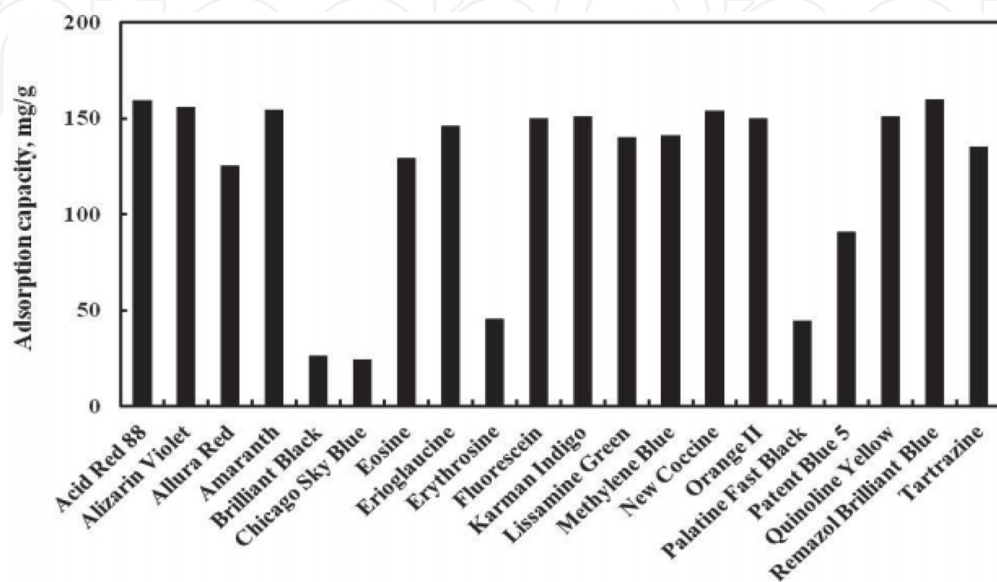


Figure 13.
 Comparison of the adsorption capacity of dyes on cationic tannin adsorbent [45].

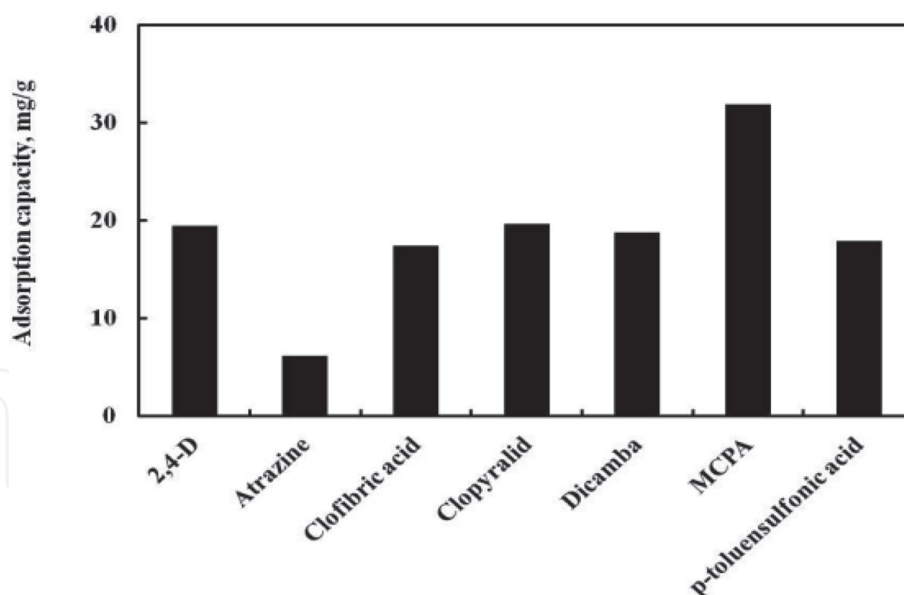


Figure 14. Comparison of the adsorption capacity of pesticides on cationic tannin adsorbent [45].

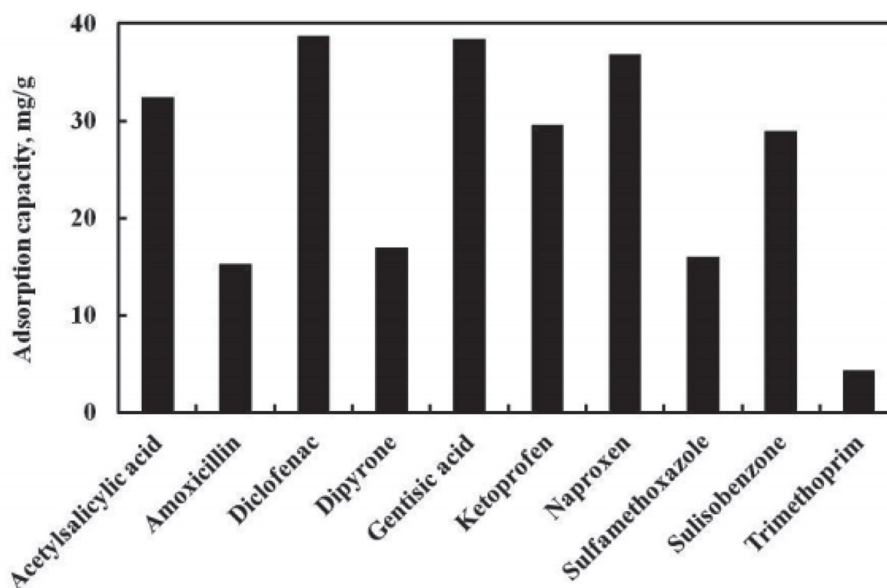


Figure 15. Comparison of the adsorption capacity of pharmaceuticals on cationic tannin adsorbent [45].

2.2.2.4 Metal protective agents

Tannins have been used as a corrosion inhibitor for carbon steel and copper in oil and gas facilities with treated *Rhizopora mucronata* tannin. Sulfonated tannins can be used to remove scale-forming cooling water pipes and boilers. In the protective coating industry, tannins have been used for protecting metal surfaces as a primer/undercoat/topcoat. In Chile, pine tannin has been used as a commercial anticorrosive product. Black wattle extract has been used as a polyurethane-type coating for wood [11].

2.2.2.5 Building construction purpose

Tannins are used as a viscosity modifier of mud for the production of residential and architectural bricks [11].

2.2.2.6 *Eco-friendly preservatives*

Generally, condensed tannins preserve lignocellulosic materials naturally. Therefore, condensed tannins can be used as potential wood preservatives and biocides [46].

2.2.2.7 *Medical science*

Tannins have antiviral, anticancer, antibacterial, anti-inflammatory, and antioxidant properties, and these are the driving forces to be a potential source of medicinal products. A large amount of research is being undertaken currently to develop medicine from tannin [11].

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