

Review:**Tropical Tannin for Engineering Application**Nor Adzwa Binti Rosli¹, Wan Asma Ibrahim², Zulkafli Hassan^{1,*}, and Azizul Helmi Bin Sofian¹¹Faculty of Chemical and Natural Resources Engineering, Universiti Malaysia Pahang, Lebuhraya Tun Razak 26300, Gambang, Pahang, Malaysia²Forest Research Institute Malaysia (FRIM), 52109 Kepong, Selangor Darul Ehsan, Malaysia*** Corresponding author:**

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Abstract: In this study, some approaches have been proposed to establish an alternative and option of brand-new compounds by using green sources that can minimize the environmental threat in the engineering application industry. Tannin, a chemical component extracted from plant origin, has the potential to bind with proteins and other polymers. The description of tannin can be amplified to cover a complete mass of constituents which give typical phenolic reactions, and hence, it has the properties to interact with the aqueous solution. The potential of tannin to associate allows its usability in the oil and gas industry. The aim of this review in this particular context will be emphasized the use of tannin in the implementation of drilling fluid, mercury removal, wastewater treatment, and corrosion inhibitor.

Keywords: tannin; corrosion inhibition; drilling fluid; mercury; wastewater

■ INTRODUCTION

Tannins in actuality are a substantial and diverse family of the phenolic compound with different structure and widely distributed through the plant kingdom. It has myriad supplemental roles in a plant's life cycle, as protective tissue and defense strategies [1]. The phenolic compound is consist of a hydroxyl group (-OH) bonded to an aromatic compound and has the properties to interact with the aqueous solution. The phenolic compound is an active compound from the lower bond separation energies of -OH group and not difficult to lose H atom [2]. Hence, tannin-containing enough hydroxyl group to encouraging effective cross-linking of other compounds. Phenol which contains -OH group attached to a hydrocarbon is a type of weak acid, and even if it is still a weak acid, phenol is sufficiently recognizably acidic properties. A hydrogen base ion can transfer to a base when it breaks away from the -OH group.

The essential property of tannin is its ability to combine with proteins and other polymers [3]. Tannin mostly found in the tree bark, galls, legumes, fruits, and vegetable, and it has the crucial property which is to

combine with proteins and other polymers. Thus, it promotes its consumption in a variety of industry such as in medicine, leather processing, dye industry, ink manufacture, and especially in engineering industries. Tannin has chemical properties such as precipitation, anti-oxidant, astringent, carcinogenicity, and reaction with salts. They have a dark brown or reddish brown color and the puckering taste. In the market, tannin can be found in powder and liquid form. It is soluble in water, alcohol, dilute alkali, glycerol, and acetone [4].

Older than the Amazon jungle and around 130 million years old ago, Malaysia's rainforests take part in some of the most various tropical ecosystems in the world. Through the Ice Age, Southeast Asia's rainforests pursue to encounter moist temperatures because of its geographical position and place. Furthermore, it allowed them to evolve much advanced than the tropical rainforests of South America and Africa. In Malaysia, there is an entire of 112 tropical mangrove forest reserves and form a segment of the country's Permanent Forest Estate which is supervised for sustainable forestry manufacture. The roots of the mangrove tree carry a

massive load of tannins, and in the soil solution, it interacts with ferric ion causing in the blackening of the roots as the tannin-ferric iron complex formed. Tannin can be extracted from mangroves species of the family *Rhizophoraceae*, which are particularly favored for making charcoal as it claimed by some to produce the best charcoal in the world [5]. The abundance of studies and productions used mangrove trees to yield tannin due to the high-quality content compared to the non-tropical tannin that extracts from other sources like a mimosa, chestnut, or quebracho trees. Studies have proven that the rich content of raw tannins up to 29% (w/w) could be extracted from the mangrove bark based on the 3-day solid-liquid extraction with 70% (v/v) aqueous acetone. Mangrove bark is inexpensive and copious available. The extraction of tannins from the mangrove bark requires simple and economical methods; hence, they are of significant commercial value.

Tannin commonly is separated into significant groups, i.e., condensed tannin, and hydrolyzable tannin. Hydrolyzable tannins can deteriorate in water, with which they react to make different substances, produce various water-soluble manufacture and products, such as gallic acid, sugars protocatechuic acid. Gallotannin, or usual tannic acid, is the finest known of the hydrolyzable tannins. The center of a hydrolyzable tannin contains a carbohydrate. The carbohydrate that contains hydroxyl groups can be classified as entirely esterified with phenolic acids gallic or partially. Hydrolyzable tannins able hydrolyzed by weak bases of weak acids to create phenolic acids and carbohydrate. Condensed tannins, recognized as proanthocyanidins, are broadly disseminated in plants and greatly influence the standard of foods. About 2–50 (or more) flavonoid units of polymers, which are not capable of hydrolysis. Some tannins which are under the group of hydrolyzable and few condensed tannins are soluble in water while massive condensed tannins are indissoluble or insoluble [6].

■ APPLICATIONS OF TANNIN

Binding Activity of Tannin

This section explains the general chemical properties of tannin in binding with certain substances. Tannins

supply hemostatic properties and astringent to a compound. They intercommunicate and interact with carbohydrates, proteins (enzyme), polysaccharides, and cell membranes of bacteria. When tannin reacts with carbohydrates, the greater the molecular weight of the carbohydrate, the more it gets across with tannins. For bonding to take place between protein and tannin, protein needs to have an open and flexible structure, a large molecular size, and to be loaded in proline. Hence, tannin requires a high molecular weight and free structures. The mechanism of bonding happens when hydrophobic and hydrogen joining hydrolyzable tannins and proanthocyanins from tannin-protein complexes in comparative ways. These groups are extremely steady structures and are exceptionally not easy for the gut to separate. It is workable for proteins to interact with tannins when the measure of protein is in overabundance or excess of that of the tannin. At the point when this happens, there is no precipitation, and it is hard to see whether the reaction has occurred. Insoluble complexes will be formed when the measure of tannins surpasses that of protein, and a hydrophobic external layer is framed [7]. The hydroxyl groups are incredible at building hydrogen bonds with proteins; the hydrophobic type of the benzene rings interface particularly firmly with proline residues in proteins; and in light of the fact that a single tannin molecule is so expansive, it can gather numerous proteins together, making the entire mass precipitated. By the facts, tannins are believed to have strong ability and tendency to bind with chemical substances, and it has been used in various usability in engineering applications.

Tannin in General Engineering Applications

Tannin is a vital additive in the manufacture of tanning leather. Tanbark from mimosa, oak, quebracho

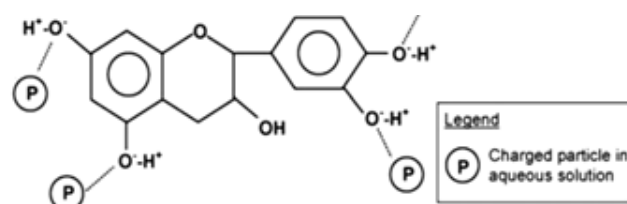


Fig 1. Representation of basic tannin structure in aqueous solution and possible molecular interaction [8]

tree, and chestnut has commonly been the main deviation of tannery tannin; nevertheless, inorganic tanning agents were also used in nowadays and consumed the world's leather manufacture for 90% [9]. Tannin or tannic acid contributes to the application in the dye industry, water purification, ink manufacture, surface coatings, plastic resins, production of adhesives, drilling fluid, removal of mercury, wastewater treatment, manufacture of gallic acid, etc. [3]. Table 1 summarizes the analysis of the usage of tannin in numerous applications by the researchers. Besides, it has been more than four decades in which tannin used to shelter and fortify steam boilers by notable levels above ASME direction. By using tannin from the green foundation as corrosion inhibitors, it minimizes the water and energy consumption, contaminants in effluent wastewaters, greenhouse gases discharge, and even decreasing the environmental mark of industrial operations [10].

Tannin as Corrosion Inhibitor

Corrosion activity is accountable for abundant losses, especially in the industrial sector. Among the diverse methods to prevent or avoid the destruction of the metal surface, the corrosion inhibitor is one of the better methods of corrosion protection and one of the most contribution in the industry. This method is considered to be good due to practice method and low cost. Crucial researches have been conducted with government investment specifically in large areas such as evolution construction of new pipelines for growth and shale gas in

construction. The attention of these researches has been the inhibitors utilization in water and concrete for the protection of metals [12]. Several papers have submitted and described the use of natural products in the growth of productive environmental corrosion inhibitors for dissimilar metals in different systems. Tannins are group of non-toxic biodegradable organic compounds, and also a polyphenol that can be built from the source of bark trees such as mimosa, quebracho, chestnut, and mangrove [13]. In the previous study, the corrosion activity in inhibited hydrochloric acid of mild steel electrodes is narrated. Typical weight loss evaluation reveals that the greatest concentration of 140 ppm of tannin from *Rhizophora racemosa* is needed to get a percentage of 72% corrosion inhibition. Natural atmospheric exposure research displayed that specimens interact in phosphoric acid H_3PO_4 , an inorganic corrosion inhibitor withstand corrosion for three weeks, and while tannin treated specimens effected with corrosion attack of experiment nearest to one week [14]. Table 2 shows the study of the different type of tannin from mangrove species as inhibitor used in corrosion test with specific specimens used in the experiment.

Fig. 2 explains the efficiency of phosphoric acid, tannin/phosphoric acid and tannin respectively in 1 M hydrochloric acid (HCl), where it is noticed that tannin presented the greatest efficiency at 140 ppm of 72% at a concentration while efficiencies of about 61 and 55% were resulted by tannin/ H_3PO_4 and H_3PO_4 respectively at

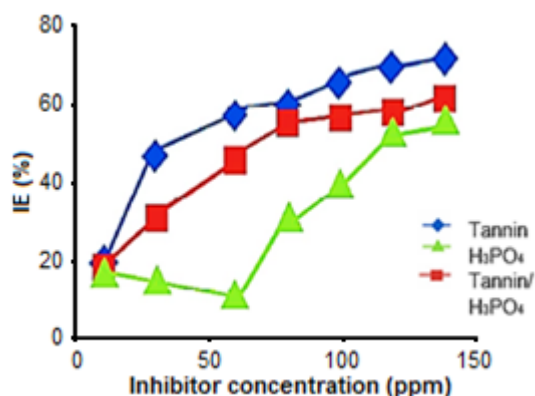
Table 1. Analysis of the use of tannin in general engineering application

Use of tannin	Engineering Application	Reference
Manufacture of gallic acid, ink manufacture, dye industry, surface coatings	Chemical, material	[11]
Possible 1 sources of new natural drugs, insecticides, herbicides, and antibiotics	Medical, environmental	[1]
Dyes, perfumes, fibers, flavoring agents, glues, oils	The material, mechanical, aromatic	[1]
Corrosion inhibitor	Material, mechanical	[12-20]
Removal of mercury	Wastewater, environmental	[21-29]
Coagulant in water treatment	Wastewater, environmental	[30-33]
Thinner in drilling fluid	Oil and gas	[34-36]
Resin	Material, mechanical	[37-40]

Table 2. Analysis of the different type of tannin as inhibitor used in corrosion test

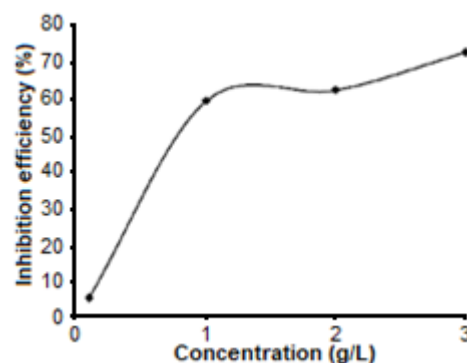
Inhibitor	Specimen	Reference
Tannin Takaot galls (<i>Tamarix articulata</i>)	Copper	[13]
Tannin bark <i>Melaleuca cajuputi</i> Powell (Gelum)	Mild Steel	[15]
Tannin <i>Rhizophora apiculata</i> mangrove bark	Mild steel	[16]
Tannin <i>Rhizophora apiculata</i> mangrove bark	Mild Steel	[17]
Tannin <i>Rhizophora racemosa</i>	Mild Steel	[14]
Tannin <i>Rhizophora apiculata</i> mangrove bark	Steel	[18]
Black wattle tannin	Carbon Steel	[19]
Modified nonionic tannic acid	Carbon Steel	[20]

the same concentration. Tannin and tannin/ H_3PO_4 efficiencies increased with greater in concentration or, in other words with the discoveries of different analysts who utilized natural based compounds as inhibitors for mild steel in different concentrations [41]. Fig. 3 shows the graph of % inhibition efficiency (IE) in the addition of mangrove tannin (MT). The graph shows the addition of % IE from 0.1 to 1.0 g/L MT followed by a gradual increase from 1.0 to 3.0 g/L MT and the IE in the presence of MT increased up to 72.8% at 3 g/L [13]. Tannin also applied in the seawater solution, and it illustrates the trend of increasing inhibition efficiency as tannin content became greater in the paint, as demonstrated in Fig. 4 with 83.86% at a higher amount of tannin (6 g) [16]. As represented in Fig. 5, tannin extracted from *Melaleuca cajuputi* Powell (gelam) added in the acidic medium shows increasing inhibition efficiency from 6 to 40.77% when the concentration of inhibitor increases from 0 to 300 ppm [15].

**Fig 2.** The corrosion of mild steel in 1 M HCl for the efficiency of various inhibitors on at 30 °C [14]

Use of Tannin as Natural Coagulant for Water Treatment

Coagulation is a chemical pre-treatment activity planned to take out colloidal substances in charge of turbidity. The sedimentation trouble of colloidal particles under the process of their own responsibility is because of the electrical charges of equal sign they bring, which disseminate repulsion among the particles by means of electrostatic forces [30]. These electrical charges are partially or totally neutralized upon coagulation. Hence, under gravitational forces, the particles can get at the same instant giving rise to great agglomerates, the flakes, which drain automatically. Most of the coagulant used in conventional treatment involves inorganic coagulants that may synthetic or plant origin as it aspiring of significant environmentally friendly behavior and natural low cost. At a recent time, a tannin-based coagulant used in water purification for

**Fig 3.** Graph of inhibition efficiency of 0.5 M hydrochloric acid with a different concentration of tannin in the presence of mangrove tannin [13]

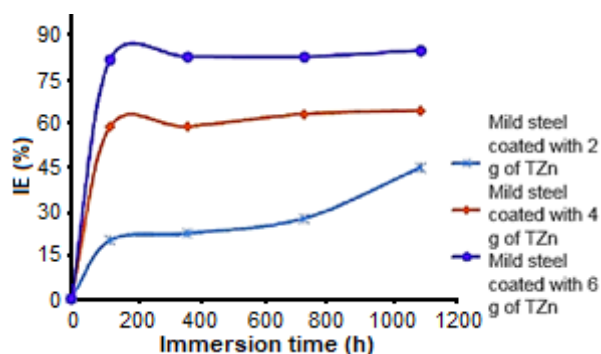


Fig 4. Inhibition efficiency of zinc tannate (TZn) at different amount during immersion in 3.1 wt.% solution in 2160 h [17]

flocculation processes. Tannin promotes amphoteric nature from the phenolic groups and does not undergo hydrolysis in solution; hence, its operational as a coagulant is consistently optimal. The benefit of tannin demonstrated as it does not change the pH of the suspension and the formation of flakes. The irregular shape supplies large surface contact, which magnifies the efficiency of flocculation [31]. The activity of tannin with aminoethanol and formaldehyde build a weakly, basic polymer that is extra successful than alum in discard turbidity and mainly color from river water. Özacar and Şengil [32] deliberate the outcome of tannin on the filterability of sludge established in the coagulation reaction and resulted that the formed sludge could be filtered as a coagulant when tannin was used. Diverse experiments that have been led on water treatment coagulant using the tannins have shown that the effectiveness of tannins is contingent on especially on the compound structure of tannins that have been extricated from the plant and degree of tannin alteration [33].

Tannin in Drilling Fluid Performance

In the process of water-based muds and drilling fluids, the thinning agent is significant to reduce the viscosity and plays a role in regulating rheological properties. These greatly effectual products avoid the need for mud dilution, which also needs replenishment of mud chemicals and grows mud volume and disposal costs. The function of thinner will preserve disseminated drilling fluid rheology; hence, securing against mud gelling or flocculation due to contamination and or high

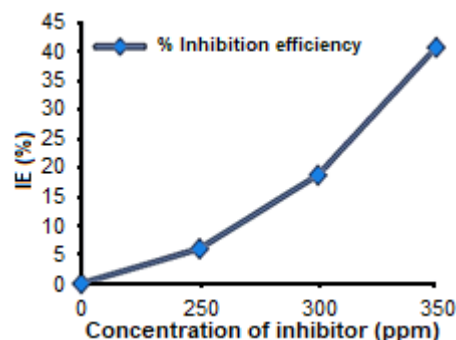


Fig 5. Inhibition efficiency of mangrove tannin in 1 M hydrochloric acid with a different concentration of tannin in the presence [15]

temperature. A thin filter-cake at the wellbore evaded associated issues, and excess fluid can be achieved in the present and help of thinners [34]. Tannins have the mechanism in which it works by thinning the clays, where the positive edges of the clay platelets are adsorbing. The reaction creates to repulsion among the platelets and plugging action of the particles themselves. The role of tannin as additive enhances the handling of solids inside the drilling fluid by deflocculating solids like bentonite and between the colloidal nature of the chemical, skewing the particle size dispersal to lower valves [35]. In a study, a chrome-free drilling mud thinner known as tin-tannin-lignosulfonate (TTLS) has been productively formulated by crosslinking tannin with lignosulfonate acid in addition of formaldehyde and stannous ions. As a result, it shows a great prospect to be commercialized from the striking performance of TTLS as a water-based drilling mud thinner. Furthermore, it is a good tolerance to salt contamination and favorable tolerance to temperature [36].

Removal of Mercury from Solution by Tannin Adsorbent

In industrial chemicals, most of the electrical and electronic application manufactured by mercury, one of the heavy metal elements that is toxic to the environment. Some other instances in an industry that consumes mercury in their process are cement industry, mining, nuclear energy production, steel production, and corrosive paints [21]. There had been issues where the waste of heavy metal are not appropriately arranged

when they are disposed of to the environment. The global development trend towards gradually of mercury-added products will come out in the generation of excess mercury if mercury provides the stand and remain at the current level. For that reason, an environmentally sound organization and management of mercury wastes will be a serious issue for nearly all countries. During previous years, there was sprouting engrossment the use of biomaterials for the preconcentration with sorption of heavy metals from water. Plant extraction as bark, algae, roots, and leaves was used to this occasion. To be specific, the waste of wood industry comes from the bark has the ability source for the recuperation of heavy metals, as it carries a huge amount of vegetable tannin [22]. The ability of tannin to forms chelates with metals because of the hydroxyl groups in its molecule become attention in studies adsorption of mercury and methylmercury in

solutions. The hydroxyls are the fundamental of useful gatherings exist in tannin molecules, and the adsorption of cations in tannin resin can be clarified by the metal chelation with adjoining hydroxyl groups of the tannin phenolic molecules. [23]. Table 3 synopsis different methods and tannin sources in the process of removing mercury in solutions.

A study on Hg(II) removal from aqueous solution by bayberry tannin-immobilized collagen fiber (BCITCF) had been proved that the adsorption of BCITCF follows the chelating process activity. The BCITCF can be simply produced with 0.1 M lactic acid following adsorption operation and recycled no less than 4 times without the loss of adsorption capacity. To sum up, the BCITCF can be utilized as a beneficial adsorbent for productive removal of mercury from aqueous solution [24].

Table 3. Analysis of the methods of removing mercury and heavy metal by tannin

Type of tannin	Methods	References
Removal of mercury(II) and methylmercury from solution by tannin adsorbents	Adsorbent synthesis of tannin resin process from <i>Eucalyptus saligna</i> Sm (ETS) and <i>Lysiilona latisiliqua</i> (LTS) in removing mercury (II) and methylmercury	[25]
Removal of cadmium and mercury ions from aqueous solution by sorption on treated <i>Pinus pinaster</i> bark: kinetics and isotherms	Formaldehyde pre-treated <i>Pinus pinaster</i> bark was used to absorb Hg ²⁺ and Cd ²⁺ from aqueous solutions	[26]
Hg(II) removal from aqueous solution by bayberry tannin-immobilized collagen fiber	Adsorption process used by immobilizing bayberry tannin (BT) onto collagen fiber was found to be effective to eliminate mercury	[24]
Biosorption of Pb(II) ions by modified quebracho tannin resin	Fourier transform infrared spectroscopy (FTIR), and Scanning electron microscopy (SEM) was used to study quebracho tannin resin on the adsorption of lead	[27]
Synthesis and characterization of tannin-immobilized hydrotalcite as a potential adsorbent of heavy metal ions in effluent treatments	An adsorbent in the removal of heavy metal ions by immobilizing tannin of modification of calcined hydrotalcite (HTC)	[28]
Removal of mercury from wastewater use of green adsorbents - a review	Compilation of method used such as chemical precipitation, conventional coagulation, adsorption, ion exchange and reverse osmosis in the removal operation of mercury	[29]

■ CONCLUSION

This review shows that the study on tannin for engineering applications have attracted the attention of more researchers. The nature and composition of tannin prove that the binding capability can be amplified to enhance the wastewater treatment, removing the heavy metal in the solution, refine and enhance drilling fluid operation and reduce the risk of corrosion in plant processing. Tannins have a good potential as an effective flocculating agent to be used as a natural coagulant for its ability to get rid of turbidity. Besides, the practical application of tannin as an adsorbent can be expected for the removal of mercury for its excellent desorption and adsorption behavior, highly effective, reusable adsorbent, and low cost.

Furthermore, in order to maintain a system in drilling fluid performance with the condition of a dispersed (deflocculated) state, the repulsive forces must be maximized. One of the ways to maximize the negative charges of the clay particles is by adding deflocculant and tannin had become the common chemical where it can be effective at low concentration. Local tannin as a natural inhibitor can replace the synthetic inhibitor since it acts as cathodic on a tested specimen based on studies of corrosion testing. By proving the use of tannin, conclusively tropical tannin that can be extracted from mangrove trees specifically can promote tannin as one of the vital chemical substances in engineering implementation and yet providing more job opportunities to those exceptionally living in mangrove areas.

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