The effect of tannins and pH on the corrosion of steel in wood extracts

S. L. Zelinka* and D. S. Stone

Tannins and pH are often cited as factors that affect the corrosiveness of wood yet there are few data to confirm these statements. The purpose of this paper is to systematically investigate the effect of tannins and pH on corrosion of metals in wood. Four wood species known to vary in both their pH and extractives were chosen and extracted with water. The pH, tannin content, and corrosion rate of carbon steel were determined for each extract. To test if pH and tannin were the only factors influencing corrosion, "synthetic" extracts were made by adding tannins to distilled water and adjusting the pH. At a given pH, increasing the tannin content inhibited corrosion and at a given level of tannin content, lowering the pH increased corrosion. An isocorrosion map as a function of pH and tannins was developed by using simple kinetic models to extrapolate the data. The data may be useful in estimating the relative corrosiveness of new, nonmetallic wood preservatives based upon the chemistry of naturally durable wood species.

1 Introduction

Recently there have been several papers on the corrosion of metals in wood treated with copper-based wood preservatives. Although the corrosion mechanism involves the reduction of cupric ions [1], it also depends on the wood extractives. *Kear* et al. studied the corrosion behavior of metals in solutions of wood preservatives and concluded that these solutions cannot be used to predict corrosion in solid wood [2, 3]. *Zelinka* et al. [4] found that the water extracts of treated wood were much more corrosive than dilute solutions of the wood preservative with roughly the same copper concentration. These studies suggest that wood extractives affect the corrosion of metals in treated wood; however, the literature on this topic is sparse, and the role of several extractives remains controversial.

Although a single piece of wood can contain over 700 different extractives [5], only three types of extractives have been thought to affect the corrosion of metals in contact with wood or the black liquors of wood pulp: small organic acids (acetic and formic acid), tannins (or more broadly polyphenols), and phenols with two or three adjacent hydroxyl groups (e.g., catechol and

E-mail: szelinka@fs.fed.us

D. S. Stone

pyrogallol). For solid wood, only organic acids and tannins have been mentioned in the literature; catechol (1,2-dihydroxybenzene) and pyrogallol (1,2,3-trihydroxybenze) are formed as the lignin is destroyed in the pulping process [6, 7].

1.1 Small organic acids

Although the pH of wood, a solid material, is not well defined, the water extracts of nearly all wood species are acidic [8–10]. The reason for this acidity is that in the presence of water, acetyl, and formyl groups in the hemicelluloses are hydrolyzed to form acetic and formic acid [10]. It has been shown that this process is chemical, rather than biological [9–12], and that over 90% of the acid produced is acetic [9].

The earliest papers on the corrosion of metals in wood attribute the corrosiveness of wood to its acidity. Farmer [13] lists the pH of several wood species and suggests a correlation between the most corrosive wood species and the most acidic wood species. Farmer emphasizes the corrosiveness of western red cedar (Thuja plicata) and lists the pH in the range 2.9-4.7. Bartel-Kornacka [14] studied the corrosiveness of 23 tropical wood species by placing iron in contact with water-saturated sawdust and found no difference in corrosiveness for species with a pH greater than 5; some species with a lower pH were more corrosive. Knotkova-Cermakova and Vlckova [15] cite the presence of acetic and formic acids as the only corrosive factors in wood. Smith [11] ranks 16 species of wood according to their corrosivity, lists their pH, and concludes that, "a strongly acid [sic] wood, pH less than 4.0, is potentially dangerous, and a less acid [sic] wood, pH more than 5.0, is likely to be relatively safe." This

S. L. Zelinka

USDA Forest Products Laboratory, One Gifford Pinchot Drive, Madison, WI 53726 (USA)

Department of Materials Science and Engineering, College of Engineering, University of Wisconsin, Madison, WI 53706 (USA)

concurs with the accepted knowledge that corrosion rate of carbon steel, while generally constant in the range of pH 5–9, increases rapidly with decreasing pH below pH 5 [16].

To summarize, many researchers have found correlations between the acidity of wood and its corrosiveness, and the pH is largely controlled by the formation of acetic and formic acid. However pH cannot be the only variable that affects corrosion. For example, *Bartel-Kornacka* [14] found differences in corrosion between species with the same pH; *Cynometra ananta* and *Nauclea diderrichii* both had a measured pH of 4.5, but the corrosion rate in *Cynometra* was twice as high as in *Nauclea*. Furthermore, when *Terminalia ivorensis* was extracted with a mixture of ethanol and water, it was much less corrosive. Similarly, *Arni* et al. [17] removed all of the volatile acids and phenolics from the wood and found the extracted wood was not corrosive toward mild steel.

1.2 Tannins

Tannins and other polyphenolic extractives are often mentioned in the literature as compounds that affect corrosion in wood. Polyphenols complex with metals; adjacent hydroxyl groups interact with metal ions [18–20] forming metal tannates which are insoluble and form a dark blue/purple precipitate [21]. However, there is disagreement about whether this chelation increases or decreases corrosion. Papers that examine the corrosion of metals in solid wood state that tannins increase corrosion [22–26], whereas studies on corrosion in the pulp and paper industry state that tannins are corrosion inhibitors [5, 18, 19, 27–29], although only one of these papers explicitly examined their effect [5]. In the broader corrosion literature, wood tannins are frequently used to protect metals as either a primer or as an inhibitor [20, 30–35].

Most of the research on the effect of tannins on corrosion in solid wood have examined the wear resistance of sawblades. *Krilov* and *Gref* [22] list the pH and polyphenol content for 15 different hardwoods, although all but two of the species were from the genus *Eucalyptus*. No corrosion data were reported, but a mechanism for corrosion involving the chelation of ferric ions was proposed. *Winkelmann* et al. [23, 24] claim that tannins accelerate the corrosion process by both lowering the pH and forming complexes with iron that do not adhere to the surface. *Pugsley* et al. [25, 26] examined the role of tannins in the corrosion of sawblades by measuring the strength of ceramic reinforced tool-steels exposed to a 5 mM (8.5 g/L) solution of tannic acid. The strength of all steels decreased with exposure time, and some alloys exhibited stress-corrosion cracking.

In contrast to the above studies, in the broader corrosion literature tannins are often cited as corrosion mitigators. For example, *Matamala* et al. tested tannins extracted from radiata pine (*Pinus radiata*) and black acacia (*Acacia melanoxylon*) as part of an anti-corrosive paint system and found that the tannins increased the time to failure by over 250% when compared to an equivalent paint system without the tannins [31]. Similarly, researchers have found that tannins extracted from radishes, honey, and various other plant species inhibit corrosion in acid and sodium chloride solutions [34–36].

The different behavior of tannins in these papers is most likely due to the studied application. In sawblade corrosion, it is

possible that the iron-tannates are removed from the surface due to friction or heat and the chelation results in a more active surface than normal corrosion processes, whereas in other situations, iron-tannates protect the surface. Regardless, the effect of tannins on the corrosion of metals in wood remains unclear.

1.3 Catechol and pyrogallol

Although catechol and pyrograllol are not expected to be found in large quantities in solid wood, they warrant discussion in this work because they have been well studied in pulp and paper corrosion and are chemically similar to tannins. All of these compounds form complexes with iron at adjacent hydroxyl groups. Gust and Suwalski used Mössbauer spectroscopy to study the interaction of iron with catechol, pyrogallol, gallic acid, and oak tannins [20]. They found that iron formed a complex with the same Mössbauer parameters for all compounds; i.e., the interaction of iron and the phenolic molecule only depended on whether the molecule contained hydroxyl groups in adjacent positions. Furthermore, they found no difference in the type of complex formed between catechol and pyrogallol. These similarities suggest that catechol and pyrogallol should affect corrosion in the same way as tannins; however, the literature does not support this.

Seminal work by *MacLean* and *Gardner* [18] found that catechol increased the corrosion of equipment used to pulp western red cedar. *Kannan* and *Kelly* [19] found that the critical current density for steel passivation in alkaline media was unaffected by 1,3-dihydroxybenzene and 1,4-dihydroxybenzene-but was greatly increased in the presence of catechol and pyrogallol [19]. Similarly, *Singh* et al. [27–29] examined the role of catechol on corrosion and found that it interacted synergistically with other components of the black liquor to increase corrosion.

1.4 Objectives of this research

The literature points to the importance of pH, tannins, and catechols as important factors in the corrosion of metals in wood and wood pulp. In this work, we systematically investigate the effects of tannin content and pH in extracts of four wood species exhibiting a range of both pH and tannins to see if there is a trend. These might not be the only factors affecting corrosion. If they were, "synthetic" extracts with the same pH and amount of tannin content as the wood extracts should give the same results. To test the influence of other factors, the corrosion rates of synthetic extracts were made and compared to the wood extracts.

2 Materials and methods

Four wood species known to vary across a range of acidities and quantity of extractives were investigated in this study by examining their water-extracts. White oak (*Quercus* spp.) was chosen as it is very acidic [10], one of the most corrosive solid woods [11], and is also high in tannins [37]. Elm (*Ulmus* sp.) was chosen because it is low in extractives and is one of the least acidic woods; literature values of the pH range from 6.0 to 7.2 [11, 13].

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Black locust (*Robinia pseudoacacia* – hereafter referred to as locust) was chosen because it is a naturally durable species [38, 39]. It has been shown that the extractives that protect the wood are water soluble [39] and contain ortho-phenolic groups [38] so it is therefore reasonable to expect that these extractives may also affect the corrosiveness of locust. Southern pine (*Pinus* spp.) was chosen to connect with existing work on treated wood [4]. Additionally, other researchers have noted the corrosivity of southern pine black liquor [27–29]. Elm and locust were harvested near Madison, WI and stored in a temperature but not humidity controlled room for several years before testing. The southern pine and white oak were purchased commercially and had been kiln-dried prior to testing. No effort was made to separate the heartwood from the sapwood; rather one homogenized extract was made for each wood species tested.

Corrosion of steel in water extracts was determined by the method of *Zelinka* et al. [4]. The extract was made by combining sawdust and distilled water in a 1:10 ratio by weight for one week at room temperature. After the extraction period, the sawdust was removed with vacuum filtration and the extract was stored at 1 °C until just before testing to minimize any possible biological activity. No effort was made to control particle size of the sawdust because in previous work it was found that the corrosion rate did not depend on the size of particles used to form the extract [4].

Corrosion of a 1018 steel plug (UNS G10180) was measured electrochemically. The potential was varied from -30 mV to +30 mV with respect to the open circuit potential at a scan rate of 0.166 mV/s. Prior to polarization, the open circuit potential was measured for an hour. The corrosion rate was determined by simultaneously measuring the polarization resistance and the Tafel slopes, which were ascertained from the non-linearity in the polarization resistance curve using *Mansfeld's* method [40, 41].

For each extract, the pH was measured with a double junction glass bulb electrode (Cole Parmer) in conjunction with a 5-star meter (Orion), and the amount of tannin was quantified colorimetrically with the method of Amin [42] with minor modifications. In this method, tetrazolium blue is reduced in the presence of tannins and the absorbance at 527 nm is measured. The color is compared against a calibration curve made with tannic acid, and the tannin content is expressed as an equivalent concentration of tannic acid. Two modifications were made to the method. First, tetrazolium blue solution was made to a concentration of 0.005 M and twice the volume used because it did not readily dissolve in water at 0.01 M. Second, when solutions were heated in 10 mL volumetric flasks, boiling over occurred, and the samples could not be tested quantitatively. Instead, solutions were heated in a 50 mL Erlenmeyer flask and after heating, transferred to a 10 mL volumetric flask where the total volume was brought to 10 mL with ethanol.

"Synthetic" extracts were made by first adding tannic acid to distilled water and then adjusting the pH by the addition of sodium hydroxide.

3 Results

The pH, tannin concentration, and corrosion rate (with standard error) are given in Table 1. Fortuitously, the pH of pine and oak

Table 1. Acidity, tannin content, and corrosion rate of the solutions tested

		pН	Tannins (mg/L) ^{a)}	Corrosion rate of steel (µm/year)	
				Mean	Standard error
Extract	Oak	4.3	1020	279	10
	Pine	4.5	340	334	26
	Locust	5.1	990	74	24
	Elm	6.5	40	159	33
Synthetic Extract	Oak	4.3	1000	49	2
	Pine	4.5	350	31	3
	Locust	5.1	1000	27	1
	Elm	6.5	40	33	4
Modified Extract ^{b)}	Pine	4.2	1000	285	24
	Locust	4.3	990	198	24

^{a)}Expressed as tannic acid equivalent.

^{b)}The modified pine extract was made by adding tannic acid to the pine extract until the total concentration of tannic acid of 1000 mg/L was reached. The modified locust extract was made by adding acetic acid until the pH was 4.3.

were similar, and the tannin concentrations were nearly equal in oak and locust. This allows a limited examination of the effect of pH and tannin individually while holding the other variable constant. Corrosion rates in the synthetic extracts were much lower than in the wood extracts, and they exhibited a different response to changes in pH and tannins.

The steel plug exposed to the oak and locust extracts formed a blue/black patina after the polarization resistance test. This behavior was replicated in the synthetic extracts, and small amounts of the patina could be found on the plug after it had been exposed to synthetic pine and elm extracts. Furthermore, in all the synthetic extracts, a blue/black cloud of precipitate was visible around the electrode (Fig. 1). This cloud was most visible in the synthetic oak and locust extract. The precipitate cloud may also have been present in the wood extracts but the extracts were translucent prior to testing so the precipitate cloud would not have been visible. It is assumed that the patina and precipitate were iron-tannate, since these solutions contained tannins, and iron-tannates are also blue/black in color [30–32].

Because of the similar pH between pine and oak extracts and the similar tannin content in oak and locust extracts, ancillary tests were conducted on modified extracts of pine and locust. The pine extract was modified by adding tannic acid until the total concentration was 1000 mg/L and the locust extract was acidified with acetic acid until the pH was 4.3; these modified extracts both had nominally the same pH and tannic acid concentration as the unmodified extract of oak. The properties of these extracts are given in Table 1 and the corrosion rates are shown graphically in Fig. 2.

4 Discussion

Although only four species were tested, some general trends can be observed in the data. The similarity in pH between pine and oak allow us to examine the role of tannins holding pH constant. It appears that increasing the tannins at a given pH decreases the corrosion rate; that is the tannins are acting as an inhibitor.



Figure 1. Blue-black precipitate forming on the surface of the steel plug exposed to synthetic oak extract

Interestingly, the modified pine extract, having the same concentration of tannins as the oak extract had the same corrosion rate. This suggests that the difference in corrosiveness between unmodified pine and oak is due to differences in tannin content, which is remarkable considering the complexity of wood chemistry.

Likewise, the effect of pH at a given tannin concentration can be examined by comparing the locust and oak extracts. Oak had a lower pH and a higher corrosion rate. When acetic acid was added to the locust extract until it had the same pH as the oak extract, the corrosion rate increased, but the modified locust was not as corrosive as oak, even though they had the same pH and tannin concentration. This suggests that locust may contain one or more compounds that act as corrosion inhibitors that are not present in either oak or pine.

The corrosion rates in the synthetic extract are all much lower than in the solid wood extracts, and there is a different trend across the species. This can be seen most clearly by plotting the extract corrosion rate against the synthetic extract corrosion rate (Fig. 3). From the data, there appear two possibilities. The first is that oak is an outlier and all of the synthetic extracts have a corrosion rate of roughly $30 \,\mu$ m/year. The other possibility is that pine is an outlier and there is a linear trend between the extract and the synthetic extract. In either case, there appear to be one or more chemicals in the wood extract that affect corrosion in addition to tannins and the pH.



Figure 2. Corrosion rates measured in the modified extracts. Error bars represent the standard error (uncertainty in the mean). The "modified" extracts of pine and locust have the same pH and tannin content as oak



Figure 3. Extract corrosion rate plotted against the corrosion rate in the synthetic extract

For a clearer understanding of the effects of tannin and pH on corrosion rate, it is necessary to invoke kinetic models. A general form for the dependence of corrosion rate (r) on the concentration of hydrogen ions (C_{H^+}) in acidic conditions (below pH 5) is given by

$$r = k_1 (C_{H^+})^n$$
 (1)

where *k* and *n* are constants; the corrosion rate of steel is largely independent of pH between pH 5 and 9 [16]. We have modeled this transition from a power-law dependence (C_{H^+}) to no dependence on (C_{H^+}) at pH 5 with a decaying exponential by adding unity to Eq. (1); i.e.

$$r = k_1' \left(e^{m(5-pH)} + 1 \right)$$
 (2)

It has been shown that the Langmuir isotherm can be used to describe the inhibitor efficiency of tannins as a function of concentration [36]. Rearranging terms in the Langmuir equation, the corrosion rate can be written as a function of the concentration of tannins (c) by

$$r = \frac{r_0}{1 + k_2 c} \tag{3}$$

where r_0 is the corrosion rate when there are no tannins, and k_2 is a second constant. Assuming that the effects of tannins and pH are orthogonal, it is possible to combine these models in the following way

$$r = r'_0 (1 + k' c)^{-1} \left(e^{m(5-pH)} + 1 \right)$$
(4)

where r'_0 , k', and m are fitting parameters and m > 0. Fitting the above model to our data allows us to examine more clearly the effects of tannin concentration and pH.

Figure 4 is an isocorrosion diagram created by fitting our model to the wood-extract data in Table 1. This figure was based on the following assumptions: (i) that the effects of tannin and pH are orthogonal; (ii) that the literature models for the effects of tannins and pH are valid for wood extracts; (iii) that the transition from pH dependent to pH independent corrosion occurs at pH 5 and can be modeled with a decaying exponential; and (iv) that the small data set is sufficient to fit the model.

More data would be needed for any reliable prediction given the model assumptions, the possibility of synergistic effects, the lack of agreement between real and "synthetic" extracts, and the need to show correlation between corrosion in extracts and in solid wood under outdoor exposure.

Despite its limitations, Fig. 4, may be used for visualizing the effects of tannins and pH. One possible application is in the development of new, completely non-metallic wood preservative systems. These preservatives are based on the chemistry of



Figure 4. Corrosion rates in wood extracts as a function of pH and tannin content. Labeled isocorrosion lines are in μ m/year. The diagram was generated by fitting the model in Eq. (4) to the data in Table 1

naturally durable species [43–45] and contain antioxidants, including tannins, that might complex with iron. This isocorrosion map could be used to predict the relative effects of adding tannins or adjusting the pH of these preservatives.

5 Summary and conclusions

The corrosion of steel in wood extracts was studied. Specifically, the corrosion rate was studied as a function of pH and tannin content. In the extracts, at a given level of tannins, a lower pH increases corrosion. In contrast, at a given pH, a decrease in tannin content increases corrosion. It appears that in wood extracts, tannins act as a corrosion inhibitor. This is in contradiction to findings in the literature on sawblade corrosion. The difference is most likely due to the friction and heat produced during sawing.

By combining kinetic models in the literature, an isocorrosion map was created for wood extracts as a function of pH and tannins. This map is based on limited data and it does not explain why the synthetic extracts behave differently; nevertheless, in the future with additional data such maps may be able to assess the relative effects of these chemicals when developing a new, nonmetallic preservative system.

The synthetic extracts exhibited lower corrosion rates and different relative rankings between wood species. It appears there is one or more chemicals not accounted for in this study that affect corrosion. These chemicals (or chemical) may act synergistically with tannins and pH or also may be unevenly distributed among the wood species tested.

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