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# Use of Reishi Mushroom Extract as Green Corrosion Inhibitor for Carbon Steel in Acid Media

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ABSTRACT

The use of Reishi mushroom or *Ganoderma lucidum (G. lucidum)* extract as a green corrosion inhibitor for 1018 carbon steel in 0.5 M $H_2SO_4$ has been evaluated by using weight loss tests. These tests were complemented by electronic microscopy analysis and infrared spectroscopy. Results have shown that *G. lucidum* extract acts as a good corrosion inhibitor, with its efficiency increasing with increasing the inhibitor concentration and decreasing with an increase in the testing temperature from 25 up to 60 °C. The corrosion inhibition of *G. lucidum* was found to be due to the presence of functional groups, mainly amines that are physically adsorbed on to the steel surface by following a Langmuir type of adsorption isotherm, which form a protective film on top of the steel.

## 1. Introduction

Iron and its alloys find extensive applications in industry, where they are used in a variety of service environments. Excessive corrosion attack is known to occur on metals deployed in service in aggressive environments. A significant method to protect such metals is the introduction of corrosion inhibitors that hinder the corrosion reaction and thus reduce the corrosion rate. Inorganic substances such as phosphates, chromates, silicates, borates, tungstates, molybdates and arsenates have been found effective as inhibitors of metal corrosion. These inhibitors have also found useful application in the formulation of primers and anti-corrosive coatings, but a major disadvantage is their toxicity and as such their use has come under severe criticism [1-8]. In general, corrosion inhibitors incorporate themselves into corrosion product films in such a way as to increase the film's capacity to prevent corrosion. Nowadays, according to the art of green chemistry, the research and development of nontoxic, natural and environmentally friendly inhibitors are carried out in a continuous mode to control the corrosion phenomenon of steel in acidic media [9-16]. The inhibitive characteristics of such compounds derive from the adsorption ability of their molecules, which contain compounds such as tannins, alkaloids, and amino acids, among others, with heteroatoms such as S, N, C, etc., with the polar group acting as the reaction center for the adsorption process. The resulting adsorbed film acts as a barrier that separates the metal from the corrodent and efficiency of inhibition depends on the mechanical, structural and chemical characteristics of the adsorption layers formed under particular conditions.

*Ganoderma lucidum* (Reishi or Ling-Zhi) has been used as health-promotion supplement owing to its anti-tumor and immuno-modulating effects [17]. The activities of liver protection, hypoglycemia, and platelet-aggregation inhibition have also been demonstrated from the fruiting bodies and cultured mycelia of Reishi [18] in addition to its use for hypertension and neoplasia. Previous studies have shown that the water soluble, polysaccharide components of Reishi exhibit anti-tumor activity and reduce tumor metastasis. Polysaccharides and triterpenes are two major categories of the bioactive ingredients, but it contains also proteins, steroids, lectins and adenosine. It has been found previously that polysaccharides from *G. lucidum* exert there in vitro and in vivo anticancer effect via an immune-modulatory mechanism. Some researchers have reported that the both polysaccharides and triterpenes possess the bioactivities of antioxidation [17], hepatoprotection [18], cholesterol stasis [19], anti-hypertension [20] and inhibiting platelet aggregation [21] due to the inhibition of enzymes such as β-galactosidase, cholesterol synthase, angiotension converting enzyme, etc. Thus, due to the antioxidants properties of *G. lucidum*, the goal of this research work, is to evaluate its corrosion inhibition properties for carbon steel in sulphuric acid.

## 2. Experimental Methods

### 2.1 Testing Solution

*G. lucidum* was grown and provided by the faculty of Agro sciences in our university. It was crushed into little pieces, soaked in 2/3 ethanol+1/3 water during 21 days obtaining a solid, wich was weighted and dissolved in ethanol and used as a stock solution and used then for preparation of the desired concentrations by dilution. Used concentrations included 0, 1, 2, 3, 4, 5 and 7 mL. Each mL of inhibitor stock solution is equivalent to 0.318 g/L. As corrosive solution, 0.5 M $H_2SO_4$ was used, which prepared with analytical grade reagents.

### 2.2 Testing Material

Commercial rods of 1018 carbon steel, measuring 6.00 mm in diameter were used for this research work. For the weight loss tests, cylindrical specimens measuring 30 mm long were prepared. They were ground down to 600 grade emery paper, washed and degreased with acetone. The weight loss value per unit area, ΔW, was calculated according to:

$$\Delta W = (m_1 - m_2) / A \qquad (1)$$

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where $m_1$ is the mass of the specimen before corrosion, $m_2$ the mass of the specimen after corrosion, and $A$ the exposed area of the specimen. For the weight loss tests, inhibitor efficiency, *IE*, was calculated as follows:

$$IE\ (\%) = 100\ (\Delta W_1 - \Delta W_2)/\ \Delta W_1 \quad (2)$$

where $\Delta W_1$ is the weight loss without inhibitor, and $\Delta W_2$ the weight loss with inhibitor. Selected specimens were observed in a LEO 1450 VP scanning electron microscope (SEM) for their analysis.

### *2.3 FTIR Spectroscopic Analysis of Green Corrosion Inhibitor*

The green corrosion inhibitor as well the corrosion products were examined under FTIR analysis by using a Bruker equipment in KBr pellet in the 4500-570 $cm^{-1}$ interval. The peak values of the FTIR were recorded. Each analysis was repeated twice to detect the characteristic peaks and their functional groups.

## 3. Results and Discussion

### *3.1 Weight Loss Tests*

The effect of *G. lucidum* concentration on the weight loss for 1018 carbon steel in 0.5 M $H_2SO_4$ is shown in Fig. 1, where it can be seen that, regardless of the testing temperature, the weight loss decreases with increasing the inhibitor concentration, reaching its lowest value with the addition of 7 mL (2.226 g/L) of inhibitor. However, it can be seen that the weight loss increases with an increase in the testing temperature. Inhibitor efficiency, Fig. 2, increases with an increase in the inhibitor concentration, obtaining its highest value, 92%, with the addition of 7 mL of inhibitor, but it decreases with increasing the testing temperature. The decrease in the weight loss with an increase in the *G. lucidum* concentration is consistent with the adsorption of the organic molecules of the inhibitor on to the steel surface covering the steel, and the surface coverage increased with increasing the inhibitor concentration, forming a protective corrosion products film, which supresses the corrosion reaction.

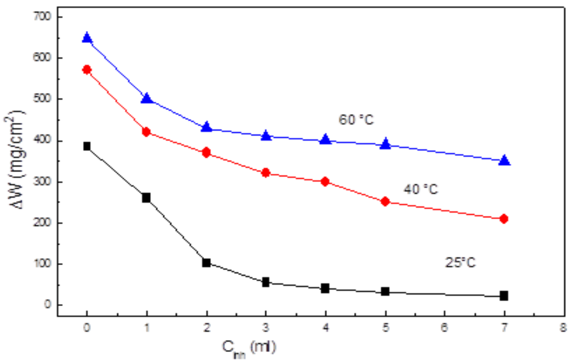


**Fig. 1** Effect of *G. lucidum* concentration on the weight loss for 1018 carbon steel in 0.5 M. $H_2SO_4$ at 25, 40 and 60 °C

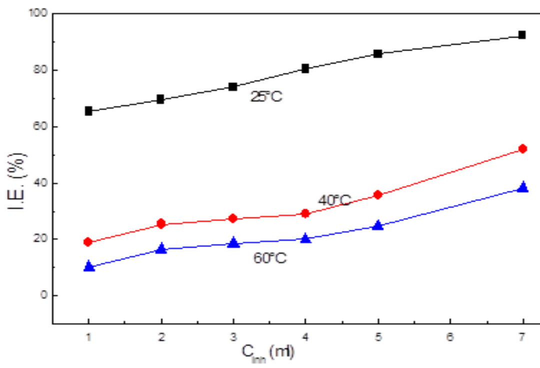


**Fig. 2** Effect of *G. lucidum* concentration on the inhibitor efficiency for 1018 carbon steel in 0.5 M. $H_2SO_4$ at 25, 40 and 60 °C

### *3.2 Adsorption Isotherms*

To investigate adsorption behaviour of *G. lucidum* extract in $H_2SO_4$ solution, numerous isotherm models were employed, such as, Langmuir, Freundlich, Flory–Huggins, Frumkin, and Temkin, but the best fit was obtained for Langmuir isotherm model (Fig. 3). This isotherm model monitors the variation of adsorption coefficient $K_{ads}$ with concentration of inhibitor $C_{inh}$ according to the following relationship [22].

$$C_{inh}/\theta = 1/\ K_{ads} + C_{inh} \quad (3)$$

where θ is the metal surface covered by the inhibitor, given by the inhibitor efficiency divided by 100, C the inhibitor concentration and $K_{ads}$ is the equilibrium constant of adsorption process. The adsorption parameters, such as, regression coefficient $R^2$, $K_{ads}$, and slope values were obtained by straight line fitting between $C_{inh}/\theta$ and $C_{inh}$, and shown in Fig. 3. As it can be seen, the $R^2$ value close to 1.0 is a verification that the Langmuir type of adsorption isotherm was a good choice. On the other hand, the $K_{ads}$ value of 3.4 L/g, and the standard free energy of adsorption for the inhibitor $\Delta G^0$ was estimated by using the equation [23]

$$\Delta G^{\circ} = -RT\ ln(55.5\ K) \quad (4)$$

gives a value for $\Delta G^0$ = -7.38 kJ $mol^{-1}$, consistent with a physical type of adsorption of *G. lucidum* onto carbon steel [24], which is associated with an electrostatic interaction between inhibitor charged molecules and charged metal surface.

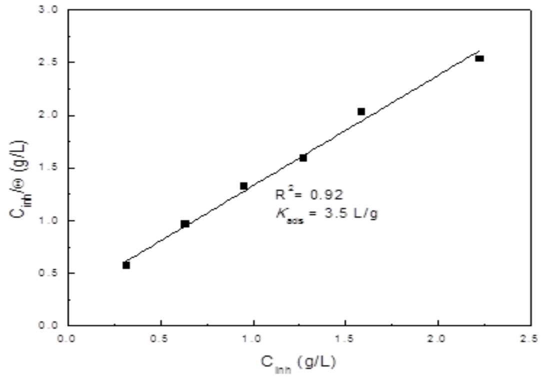


**Fig. 3** Langmuir type of adsorption isotherm for 1018 carbon steel in 0.5 M. $H_2SO_4$ at 25 °C

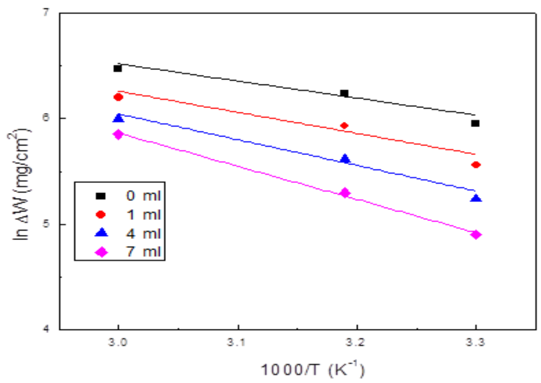


**Fig. 4** Arrhenius type of plots for carbon steel in 0.5 M. $H_2SO_4$ at different *G. lucidum* concentrations

We have seen that the dissolution of carbon steel in acidic medium increases with increase in temperature which could be due to a decrease in hydrogen evolution over potential [25]. This shows that the *G. lucidum* extract is effective in minimizing the corrosion at higher temperatures to some extent. The inhibition efficiency of the extract drops with increasing temperature. The inhibition efficiency at 25 °C was 92%, which falls to 27% at 60 °C. The decrease in the inhibition efficiency could be due to increased dissolution of carbon steel with increasing the testing temperature and desorption of the adsorbed inhibitor molecules from the metal surface or to the inhibitor degradation [26]. The apparent activation energy, $E_a$, associated with 1018 carbon steel in uninhibited and inhibited acid solution was determined by using an Arrhenius-type plot according to the following equation:

$$\ln \Delta W = -E_a / 2.303RT + \ln F \quad (6)$$

where ΔW is defined in Eq. (1), R is the molar gas constant, T is the absolute temperature and F is the frequency factor. Arrhenius plots of (in ΔW) against 1/T for 1018 carbon steel in 0.5 M $H_2SO_4$ in absence and presence of *G. lucidum* is shown in Fig. 4. The apparent activation energy obtained for the corrosion process in the free acid solution was found to be 26.1 and 76.46 kJmol$^{-1}$ in absence and with the addition of 7 mL of *G. lucidum* respectively. Notably, the energy barrier of the corrosion reaction increased in the presence of the inhibitor, which can be due to the physisorption of the inhibitor on the steel surface.

When the inhibition efficiency drops with increasing temperature and the $E_a$ in the presence of inhibitor is higher than that in the absence of inhibitor, then the adsorptive film formed on the surface of the metal is believed to be due to physical adsorption [27]. A drop in inhibition efficiency can also be explained by increased dissolution of metal at higher temperature and weakening of the physisorbed, as shown above, inhibitor layer at higher temperatures in the presence of inhibitor [28].

### *3.3 SEM Micrographs*

Fig. 5 shows the SEM micrographs of 1018 carbon steel specimens corroded in 0.5 M $H_2SO_4$ without and with 7 mL of *G. lucidum* at 25 °C with and without corrosion products. It can be seen that the film formed on the specimen exposed to the uninhibited solution contains many porous and cracks and the underlying metal suffered from severe uniform type of corrosion (Figs. 5a and c). On the other hand, the film formed on the specimen exposed to the solution containing 7 mL of *G. lucidum* does not contain porous or cracks (Fig. 5b), which makes this film more protective against the corrosive action of the electrolyte, which can be corroborated in Fig. 5d, which shows the steel surface, with evidence of much less damage due to the action of the environment. This film is the result of the reaction of $Fe^{2+}$ ions, released during the corrosion of steel, and the *G. lucidum* extract components present in the solution.

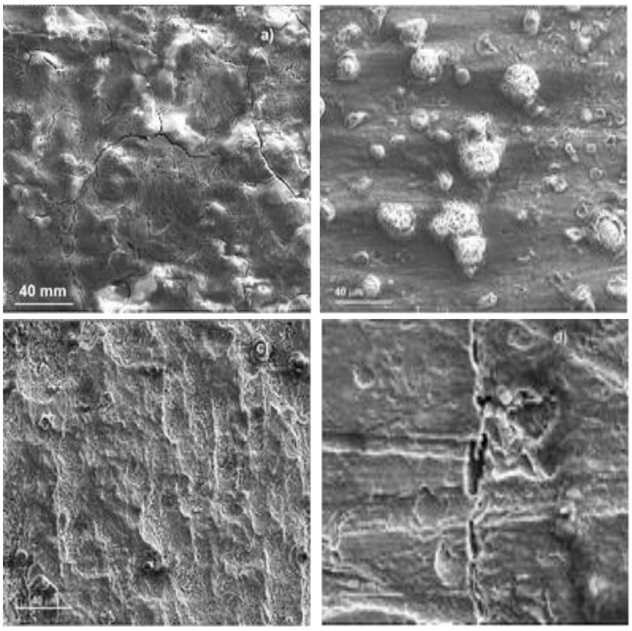


**Fig. 5** SEM micrographs of 1018 carbon steel specimens corroded at 25 °C with 0 (a and c) and 7 mL (b and d) of *G. lucidum* with (a and b) and without (c and d) corrosion products

### *3.4 FTIR Analysis*

In order to know which components there are in the *G. lucidum* extract, an FTIR analysis was performed on the extract at 25 °C, and shown in Fig. 6, whereas the FTIR spectra to the extract at 40 and 60 °C are shown in Fig. 7. For the FTIR spectrum at 25 °C, a typical signal of the O-H link, which is known to passivate metals, can be seen at 3382 cm$^{-1}$, whereas a strong signal is observed at 2978 cm$^{-1}$ which corresponds to the N-H links, very likely to the amine group; amines are known as very efficient inhibitors for iron and steel in acidic solutions [29]. The peak observed at 2900 cm$^{-1}$ corresponds to the methyl, $-CH_3$, group, whereas a strong signal at 1643 cm$^{-1}$ is for the C-O link, which is an evidence of the presence of the carboxylic acid. On the other hand, a signal at 1390 cm$^{-1}$ corresponds to the vibrations for the C-C link, characteristic for aromatic rings, and the signal at 1046 cm$^{-1}$ has been assigned to the C-N group, whereas the signal observed at 872 cm$^{-1}$ correspond to the vibrations of the C-H group. On the other hand, when the extract is heated either at 40 or 60 °C, Fig. 7, FTIR spectra indicate that the amine, methyl and C-C groups have disappeared. Maybe this fact, the absence of these functional groups, explains why the *G. lucidum* efficiency decreases when it is heated up to 40 or 60 °C. Something similar to the acid solution containing 7 mL of *G. lucidum* after the weight loss test, Fig. 8, where the FTIR spectrum indicates that the amine, methyl and C-C groups are not any longer in the solution. However, when an FTIR analysis is performed to the corrosion products formed onto the steel corroded in the solution containing 7 mL of inhibitor, Fig. 9, data show the presence of amine, methyl and C-N groups, together with Fe-O, which corresponds to some iron oxides present in the corrosion products film. Thus, all the evidence indicates that the compounds responsible for the corrosion inhibition of 1018 carbon steel in presence of *G. lucidum* contain amine, methyl and C-N groups.

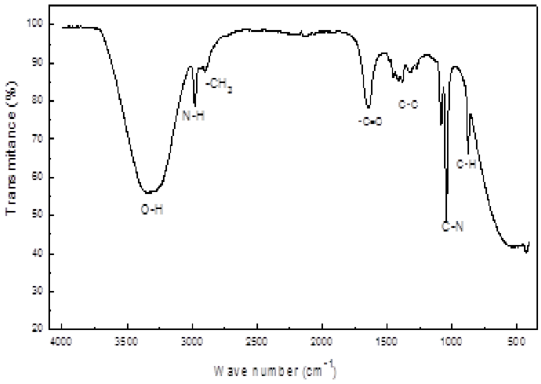


**Fig. 6** FTIR spectrum of pure *G. lucidum* extract at 25 °C

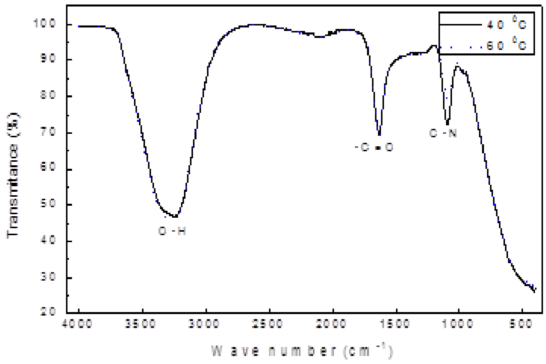


**Fig. 7** FTIR spectra of pure *G. lucidum* extract at 40 and 60 °C

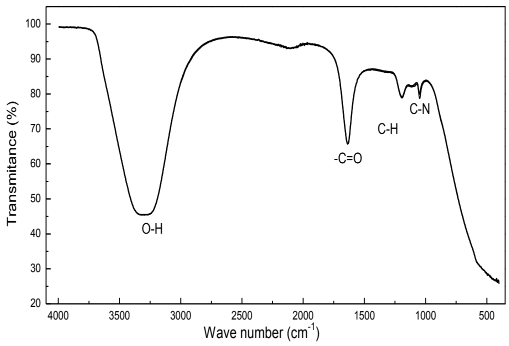


**Fig. 8** FTIR spectrum of pure *G. lucidum* extract at 25 °C after the weight loss test

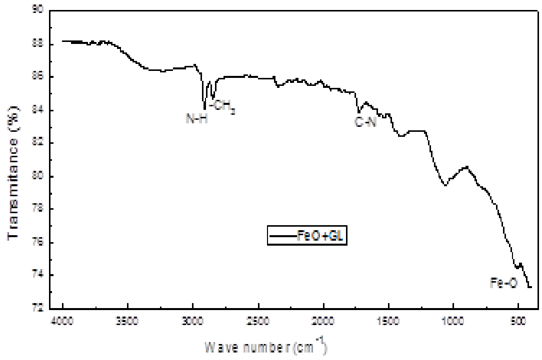


**Fig. 9** FTIR spectrum of corrosion products of 1018 carbon steel after the weight loss test in presence of 7 mL of *G. lucidum* at 25 °C

Thus, it has been proved in the *G. lucidum* extract there are functional groups such as amines and methyl groups, with heteroatoms such as C and N within them which are the responsible for the antioxidant properties of *G. lucidum.* Fouda et al [29] found that the corrosion inhibition of carbon steel in sulfuric acid in presence of aliphatic amines increased with the amine concentration, and that amines formed a complex with iron ions and amines which was physically adsorbed on to the steel following a Langmuir type af adsorption isotherm. As shown above, the interaction mechanism between *G. lucidum* and the metal involves physical interaction, i.e. electrostatic interaction between inhibitor charged molecules and charged metal surface. When carbon steel is immersed in $H_2SO_4$ solution, the steel is positively charged [30] which makes that the negatively charged $SO_4$ molecules tend to be adsorbed onto the steel surface. Additionally, it has been shown [31] that the *G. lucidum* components can be protonated, especially in its N atoms. The bonding of the *G. lucidum* derivatives on the metal surface is due to presence of lone pairs from heteroatoms (C, N, and O) and $\pi$-orbitals, blocking the active sites and therefore decreasing the corrosion rate [32-33]. Therefore, bonding between inhibitor molecules onto carbon steel surface occurs through sharing electrons of the amine group present in the *G. lucidum* extract components and vacant *d*-orbitals of iron in the 1018 steel. When this occurs, a complex between the inhibitor and $Fe^{2+}$ ions occurs covering the steel and protecting it against further corrosion.

## 4. Conclusion

A study of the possibility of using *G. lucidum* extract as corrosion inhibitor for carbon steel in 0.5 M $H_2SO_4$ has been carried out. It was found that *G. lucidum* extract is a good corrosion inhibitor for carbon steel in sulphuric acid. Inhibitor efficiency increases with increasing *G. lucidum* extract concentration but it decreases with an increase in the temperature. Corrosion inhibition is due to the presence of compounds, mainly amine group, which contain heteroatoms within their chemical structure. These compounds are physically adsorbed on to the metal surface following a Langmuir adsorption type of isotherm to form protective corrosion products.

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