



# Ginkgo biloba fruit extract as an eco-friendly corrosion inhibitor for J55 steel in CO<sub>2</sub> saturated 3.5% NaCl solution



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## ABSTRACT

The inhibition of the corrosion of J55 steel in 3.5 wt.% NaCl solution saturated with CO<sub>2</sub> by the fruit extract of *Ginkgo biloba* (GFE) has been studied using mass spectrometry (MS), Fourier transform infrared spectroscopy (FTIR), electrochemical impedance spectroscopy (EIS), potentiodynamic polarization, contact angle measurements, and scanning electron microscopy (SEM). Inhibition efficiency was found to increase with increasing concentration of the extract. The adsorption of the extract on the J55 steel surface obeyed the Langmuir adsorption isotherm. The adsorbed film on J55 steel surface containing inhibitor was confirmed by the contact angle, and SEM results. The results obtained showed that the GFE could serve as an effective inhibitor of the corrosion of J55 steel in 3.5% NaCl saturated with CO<sub>2</sub>.

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## Introduction

The total annual cost of corrosion in the oil and gas production industry is estimated to be \$1.372 billion, broken down into \$589 million in surface pipeline and facility costs, \$463 million annually in downhole tubing expenses, and another \$320 million in capital expenditures related to corrosion [1]. Various factors have to be considered while dealing with corrosion problems of oil and gas industry. Water and carbon dioxide, produced or injected for secondary recovery, can cause severe corrosion of oil well steels. Approximately 60% of oilfield failures are related to CO<sub>2</sub> corrosion mainly due to inadequate predictive capability and the poor resistance of carbon and low alloy steels to this type of corrosive attack [2,3]. CO<sub>2</sub> can produce not only general corrosion but also localized corrosion, which is a much more serious problem.

Inhibitors are chemicals that are used to mitigate the surface of metals used in oil and gas industries to mitigate corrosion. The inhibitor is adsorbed on the surface of the metal forming a compact protective thin layer [4]. Because of increasing ecological

awareness and strict environmental regulations, as well as the inevitable drive toward sustainable and environmentally benign processes, attention now has been focused toward the development of nontoxic alternatives to inorganic and organic inhibitors applied so far [5,6]. There have been many research reports on natural products as corrosion inhibitors in different aggressive environments [7–10].

Ginkgo is a widely used herbal derived from the *G. biloba* tree, a “living fossil”, being the only extant species of what was a large order of plants (*Ginkgoales*) more than 200 million years ago. Ginkgo is native to central China, but has been introduced worldwide. The word ginkgo derives from a Japanese approximation of the Chinese word for “silver apricot” referring to the tree’s fruit. Extracts from ginkgo were used in traditional Chinese medicine for centuries for a multitude of illnesses and conditions. Ginkgo extracts contain multiple compounds, but ginkgolides and bilobalide are unique to this herb. Ginkgo extracts have been shown to have antioxidant, anti-inflammatory and antihistaminic activity. Current uses are many and include dementia, memory loss, headache, dizziness, tinnitus, hearing problems, difficulty concentrating, mood disturbances, peripheral vascular disease, asthma, and bronchitis. The molecules are rich in heteroatoms (O, N, S) which are present in an effective corrosion inhibitor. Studies

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done on *G. biloba* in HCl and H<sub>2</sub>SO<sub>4</sub> on cold roll steel reveals that it can act as good inhibitor in HCl than H<sub>2</sub>SO<sub>4</sub> [11]. The influence of *G. biloba* on corrosion of carbon steels (J55, N80, P110SS and C110 steels) in 3.5 wt.% NaCl solution saturated with CO<sub>2</sub> was evaluated using static high pressure and high temperature (HPHT) autoclave [12]. The extracts of *G. biloba* leave have been investigated on the corrosion inhibition of Q235A steel with weight loss and potentiodynamic polarization techniques [13]. The inhibition of the corrosion of N80 steel in 3.5 wt.% NaCl solution saturated with CO<sub>2</sub> by *G. biloba* was investigated using electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization techniques [14]. After literature survey we concluded that no work has been done on J55 stainless steel in CO<sub>2</sub> saturated 3.5% NaCl solution with GFE. This led us to the present study to investigate the inhibiting effect of fruit extract of *G. biloba*.

## Experimental

### Preparation of ginkgo fruit extract

Ginkgo fruits were dried and ground to powder form. Dried (50 g) powder was soaked in ethanol (900 mL) and left for 24 h. Next day the ethanol was evaporated using vacuum condenser. Then the residue was mixed in 3.5% NaCl and was refluxed for 5 h. The solution was filtered and concentrated to 100 mL. This solution was used to study the corrosion inhibition properties. Corrosion tests were performed on a J55 steel of the following percentage composition (wt.%): C 0.24; Si 0.22; Mn 1.1; P 0.103; S 0.004; Cr 0.5; Ni 0.28; Mo 0.021; Cu 0.019; Fe balance, which were metallographically polished according to ASTM A262, with fine grade emery papers from 600 to 1200 grade. J55 steel coupons having dimensions of 30 mm × 3 mm × 3 mm were used for the electrochemical study. The specimens were washed thoroughly with double distilled water and finally degreased with acetone and dried at room temperature. The aggressive solution 3.5% NaCl was prepared by dilution of analytical grade NaCl with double distilled water and all experiments were carried out in unstirred solutions at room temperature.

### Mass spectrometry (MS)

The mass spectrometry was performed on GC7890–MS5975 gas chromatography mass spectroscopy (Agilent, USA) equipped with a HP-5MS capillary column (30 mm × 0.32 mm × 0.25 μm) and quadrupole mass spectrometer. Carrier gas used in the process was He (1 mL min<sup>−1</sup>). Relative component concentrations were calculated based on peak areas without using correction factors. A number of prominent peaks were identified in the repeated MS analyses of GFE. Among the peaks identified, they are very similar to D-galactose, simple phenolic acids and alkyl phenols as shown in Fig. 1.

All the peaks (Fig. 2) were matched in NIST library with their individual CAS number to get the exact information about the peaks and the results are no. 20368; 028564-83-2: 4H-pyran-4-one, 2,3-dihydro-3,5-dihydroxy-6-methyl, 100% at *m/z* 43.1, 60.4% at *m/z* 44.1, 55.1% at *m/z* 144.0, 42.4% at *m/z* 101.0, 26.8% at *m/z* 55.1. Several other peaks of other figures were identified, matched with NIST library according to their CAS number and are reported in Table S1 (Supporting information).

### Fourier transform infrared spectroscopy (FTIR)

FT-IR spectra (KBr) in transmittance mode were recorded on a Nicolet-6700 spectrophotometer. The peaks obtained as in Fig. 3 are 933.10 –OH group/olefins single substitution; 995.84 C–O bending; 1058.59 C–O stretching; 1254.66 C=C (Aromatic)/C–O stretching; 1403.67 C=C (Aromatic); 1458.57 C–H bending; 1650.72 C=C stretching; 1846.79 C–H bending; 2121.29 C≡C

stretching; 2360.49 CN stretching; 2854.59 –CH stretching vibration; 2925.17 –CH stretching/OH group.

### Electrochemical measurements

The electrochemical studies were made using an Autolab three electrode cell assembly at room temperature. The J55 steel was the working electrode, platinum electrode was used as an auxiliary electrode, and standard calomel electrode (SCE) was used as reference electrode. All electrochemical measurements were carried out using Autolab Potentiostat/Galvanostat (Model GSTAT302N) with EIS software Autolab Instruments Inc., Netherland. Autolab applications include software FRC for EIS measurements and data fitting. Prior to the electrochemical measurement, a stabilization period was allowed to attain a stable value of  $E_{\text{corr}}$ .

Tafel curves were obtained by changing the electrode potential automatically from −300 to +300 mV versus corrosion potential ( $E_{\text{corr}}$ ) at a scan rate of 1 mV s<sup>−1</sup>. Scan rate affects the linearity of the polarization curves. So, scan rate of 1 mV s<sup>−1</sup> was selected for the study, which provided the linear and smooth polarization curves. EIS measurements were carried out in a frequency range from 100 kHz to 0.00001 kHz under potentiodynamic conditions, with amplitude of 10 mV peak-to-peak. It is important that the impedance response of a system is linear. The linear condition implies that the impedance response is independent of the perturbation amplitude. This can be achieved by using small amplitude perturbations. A very small value can give rise to poor signal to noise ratio and hence noisy data. A large value can result in the violation of the linearity condition. Also, the small amplitude signals (~10 mV) superimposed on the dc potential of the interface has numerous advantages. The response is then linear function of the applied perturbation and can be entirely described by the impedance of the electrochemical system. In these conditions all the different types of perturbing signal gives the same quantity and therefore lead the same information. Typically a value of 10 mV is used for most electrochemical systems [15]. The linear Tafel segments of anodic and cathodic curves were extrapolated to corrosion potential to obtain corrosion current densities ( $I_{\text{corr}}$ ). The inhibition efficiency was evaluated from the measured  $I_{\text{corr}}$  values using the relationship:

$$\eta\% = \frac{I_{\text{corr}}^0 - I'_{\text{corr}}}{I_{\text{corr}}^0} \times 100 \quad (1)$$

where  $I_{\text{corr}}^0$  and  $I'_{\text{corr}}$  are the corrosion current in absence and in presence of inhibitor, respectively. The charge transfer resistance values were obtained from the diameter of the semi circles of the Nyquist plots. The inhibition efficiency of the inhibitor has been found out from the charge transfer resistance values using the following equation:

$$\eta\% = \frac{R'_{\text{ct}} - R_{\text{ct}}^0}{R'_{\text{ct}}} \times 100 \quad (2)$$

where  $R'_{\text{ct}}$  and  $R_{\text{ct}}^0$  are the charge transfer resistance in presence and in absence of inhibitor, respectively.

### Contact angle measurement

Contact angle measurements were performed using sessile drop technique using DSA100 Kruss optical contact angle measurement instrument made in Germany. J55 steel samples were carefully cleaned to avoid the surface contaminations, which influence the contact angle measurements through contamination of the liquid when the latter is put in to contact with the sample surface.

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