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Improvement of Corrosion Resistance for Brass in 3.5% NaCl Media by Using 4-fluorophenyl-2,5-dithiohydrazodicarbonamide

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Abstract: Corrosion poses a significant challenge for numerous industries. The use of corrosion inhibitors is essential within these industries. The efficacy of environmentally friendly corrosion inhibitors should remain high even when used at low concentrations. In the present study, the compound 4- fluorophenyl-2,5- dithiohydrazodicarbonamide (FTSC) was used as a corrosion inhibitor for brass in 3.5% NaCl solution. The inhibitor efficiency was determined by using a series of electrochemical techniques such as open circuit potential (OCP), potential dynamic polarisation (PDP), linear polarisation resistance (LPR), and electrochemical impedance spectroscopy (EIS). All experimental tests have been done in stagnant conditions. The findings of the experiments revealed that the compound FTSC looked to be of the cathodic type. Furthermore, the maximum inhibitor efficiency was reached at 98.28% at 1×10^{-3} and at an immersion time of 1 h. The current density was reduced from 16.5 to 0.284 μA.cm^{-2}. The adsorption of compound on the brass surface in 3.5% NaCl solution obeyed the Langmuir isotherm with a low negative value of the standard Gibbs free energy of adsorption (-33.8 kJ/mol ΔG_{ads} (chemisorption and physisorption). The results confirmed that the compound FTSC can be used as a corrosion inhibitor for brass in 3.5% NaCl.

Keywords: Corrosion Inhibition; Electrochemistry; Brass.

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1. INTRODUCTION

Brass, a Cu-Zn alloy, is ubiquitous due to its practical and aesthetic use across a wide range of sectors. Due to its resistance to corrosion, brass is often used in heat exchangers, construction, and maritime engineering. Brass is susceptible to dissolving in solutions with high oxygen concentrations as well as chloride, sulfate, and nitrate ions, in spite of the corrosion resistance provided by the oxide layers that have developed on its surface (1). In general, corrosion may be minimized by regulating pH levels or using inhibitors (2). Theoretically, heterocyclic organic molecules do what they do by combining with the inhibitor to make an insoluble polymeric complex. This complex then forms a protective layer on the surface of the metal. Brass corrosion inhibitors in chloride media have been researched for many kinds of compounds (3,4).

Inhibitors have been found to be the most effective approach for protecting metals from alkaline corrosion in recent years, outperforming other widely used techniques including cathodic and anodic protection, alloying, and coating (5). On metal surfaces, organic inhibitors are often used to provide a light barrier. Organic compounds containing highly electronic heteroatoms (P, S, N, and O) or heteroatoms with an aromatic ring are the most effective (6,7). Furthermore, pi electron-containing compounds with functional groups such as –N=N, -C=N, and ˃NH often exhibit corrosion properties (8,9). N- and S-heterocyclic compounds are the best corrosion inhibitors for copper in alkaline environments (10–12). Organic chemicals have a long history and are by far the most advanced and economical tool employed by manufacturers. The majority, however, are toxic and detrimental to the ecology. Global attempts have been made to find a cheap, non-hazardous corrosion inhibitor that is also effective (13–15).

The compound 4-fluorophenyl-2,5-dithiohydrazodicarbonamide (FTSC) was synthesized and investigated in our previous work as a corrosion inhibitor for mild steel (or copper) in acidic (or alkaline) solution with excellent results (10,12). Therefore, in this work, we used a corrosion inhibitor for brass in a 3.5% NaCl solution. The evaluation of inhibitors has been measured by electrochemical techniques such as open circuit potential (OCP), linear polarization resistance, potentiodynamic polarisation, and electrochemical impedance spectroscopy (EIS).

2. MATERIALS AND METHODS

Brass rod has been bought from a local market with the following chemical composition (%Wt): 58% Cu, 40% Zn, and 2% Pb. Cylindrical specimens having a diameter of 0.8 cm and a length of 3 cm were prepared from brass rod. The brass has been used to investigate the corrosion resistance of 3.5% NaCl without and with the presence of the corrosion inhibitor FTSC. The sample was connected from the back by copper wire, and after that, it was put in epoxy resin with an exposed surface area of 0.502 cm^2. Specimens were polished using silicon carbide papers, starting from 600 up to 2500 grits, to acquire a mirror-like finish. After polishing, specimens were thoroughly washed with double-distilled water and dried in the air.

All electrochemical measurements were performed using an electrochemical analyzer of type COMPACTSTAT (IVIUM). Potential dynamic polarization (PDP), linear polarization resistance (LPR) measurements of brass samples immersed in 1 M HCl solution without and within corrosion inhibitors were made at a scan rate of 60 mV/min at room temperature. The potentials were starting from a cathodic potential (-0.5 V) against a corrosion potential (Ecorr) and being allowed to sweep in the anodic direction till (0.1 V) above the Ecorr and the potential scan was reversed down to a potential equal to Ecorr. LPR tests were carried out at a range ± 10 mV with respect to OCP. The electrochemical impedance spectroscopy (EIS) technique was used to evaluate the corrosion behavior of brass samples with and without corrosion inhibitors. The experiments were performed at room temperature over the frequency range between 100 kHz and 0.01 Hz at open circuit potential. The amplitude of the voltage signal was 5 mV. The system corrosion cell was made from beaker glass of size 100 mL, the brass sample with 0.502 cm^2 was used as a working electrode, a platinum wire was used as a counter electrode; and silver chloride (Ag/AgCl) was used as a reference electrode. All plots were fitted to 1 cm^2.

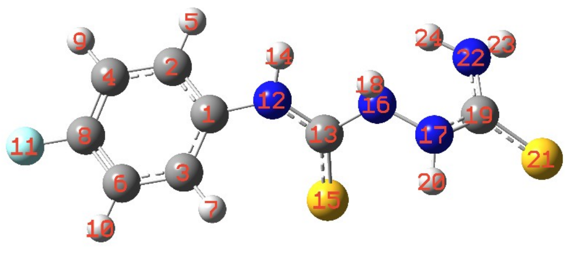

Figure 1: 4- Fluorophenyl-2,5-dithiohydrazodicarbonamide (FTSC) structure (10).

3. RESULTS AND DISCUSSION

3.1. OCP, PDP and LPR Studies

First, the open circuit potential (OCP) of brass was measured for 60 minutes in both a blank solution and in the presence of an inhibitor**.** The obtained results are shown in Fig. (2) for compound FTSC. At the start of immersion in the inhibitor-free solution, the OCP value swings in the negative direction, which might be related to the creation of an oxide film on the brass surface. The OCP values get increasingly positive as the immersion duration increases, suggesting that the oxide film is dissolving and a Cu/Zn chloride layer is forming (16–18). When an inhibitor is present, the potential at the end of the test moves in a more negative direction. This phenomenon might be due to the inhibitor molecules adsorbing on the active sites of brass and forming a protective layer (19–21).

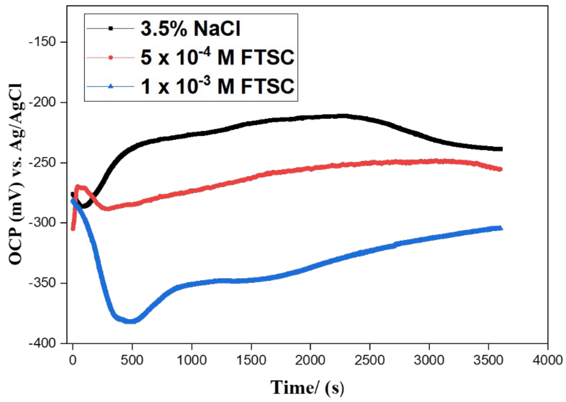

Figure 2: Open circuit potential for brass in 3.5% NaCl solution with and without the presence of FTSC.

The potentiodynamics polarization and linear polarization resistance measurements were used to explore corrosion inhibitor. Corrosion potential (Ecorr), anodic (β_a) and cathodic ($-\beta_c$) Tafel slopes, corrosion current density (I_{Corr}), surface coverage (θ), Resistance polarization (R_{pl}) and inhibition efficiency (IE%) all kinetic corrosion values were calculated and listed in Table (1) for FTSC. The IE_{PDP} % and IE_{LPR}% values were calculated by using the following equations (22,23):

$$IE_{PDP}(\%)=\frac{i^{\circ}_{corr}-i^{\circ}_{corr(inh)}}{i^{\circ}_{corr}}\times 100 \quad (1)$$

$$IE_{LPR}(\%)=\frac{R_{P(inh)}-R^{\circ}_{P}}{R_{P(inh)}}\times 100 \quad (2)$$

Figure (3) shows Tafel curves for unihibited brass and inhibited brass with the presence of different concentrations of FTSC in 3.5% NaCl at immersion time of 1 h. It can be seen that both anodic and cathodic curves changed remarkably leading to reduce the corrosion current with increase the concentration of corrosion inhibitor due to adsorption molecules on the active part of brass that lead to protect brass surface to ingress of Cl^- ions. Moreover, it can be seen from Figure 3 for FTSC, Corrosion potential (E_{corr}) was significantly shifted from -0.221 V without corrosion inhibitor to -0.329, and -0.251 V with the presence of corrosion inhibitor FTSC at concentration 5 × 10^{-4} M, and 1 × 10^{-3} M, respectively due to adsorb the molecules of inhibitor on the surface as results of increasing amount of inhibitors FTSC. Furthermore, the current density was reduced from 16.5 µA.cm^{-2} without corrosion inhibitor to 0.284 µA.cm^{-2} with presence of corrosion inhibitor FTSC at concentration 1 × 10^{-3} M due to increase the area of adsorption for inhibitor compound on the surface of brass and prevent the surface from aggressive ions. In other words, the corrosion inhibitor efficiency was increased systematically from 97.51% at low concentration of inhibitor to 98.28% at high concentration of inhibitor FTSC. The resistance polarization for compounds was approved the Tafel results. The corrosion inhibitor can be classified as cathodic type inhibitor due to move E_{Corr} to higher than 0.085 V. The maximum displacement in the E_{Corr} with presence inhibitors was 0.108 V (23).

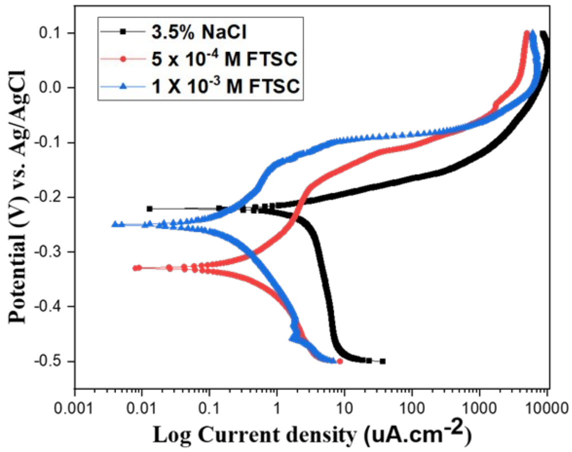

Figure 3: PDP for brass in 3.55% NaCl solution with and without the presence of FTSC.

3.2. Electrochemical impedance studies (EIS)

All parameters for electrochemical impedance spectroscopy calculated from equivalent circuit as shown in Figure.5 fitting and listed in Table.2 for compound FTSC. The corrosion inhibitor efficiency was calculated by using equation as shown below (24):

$$IE_{EIS}(\%)=\frac{R_{P(inh)}-R_P^{\circ}}{R_{P(inh)}}\times 100 \quad (3)$$

Figure 4 (A-C) illustrate Nyquist and Bode curves for compound FTSC. It can be noticed that from Figure (4), Nyquist curve for brass without inhibitor contain fading semicircle in the high frequency which is referred to the roughness and inhomogeneity of electrode , while at low frequency the shape of plot is changed to straight line due to the diffusion of soluble brass species from brass surface to bulk solution. In addition, Figure 4 also depicts that it can be noticed that the diameter of semicircle turns into bigger with presence of corrosion inhibitor due to protect the surface by molecules of corrosion inhibitor (25). It is well known that the impedance |Z| at lower frequencies the parameter utilized for evaluating the inhibitor's corrosion resistance. Figure (B-C) shows that in comparison to uninhibited brass, the impedance |Z| has increased as the inhibitor concentration has increased, indicating the robustness of inhibitor barriers to corrosive media. With FTSC present, the curve of the phase angle becomes significantly higher and it reached 75°, as seen in Figure 4 (C) .The cause for this behavior might be attributed to the adsorption of the FTSC molecule relaxing effect (12). The EIS results are in agreement with OCP, LPR, and PDP measurements. Figure 5 shows equivalent circuit for brass without inhibitor FTSC and with the presence of inhibitor FTSC. Rs is resistance of solution, CPE is constant phase element, and Rp is resistance polarization.

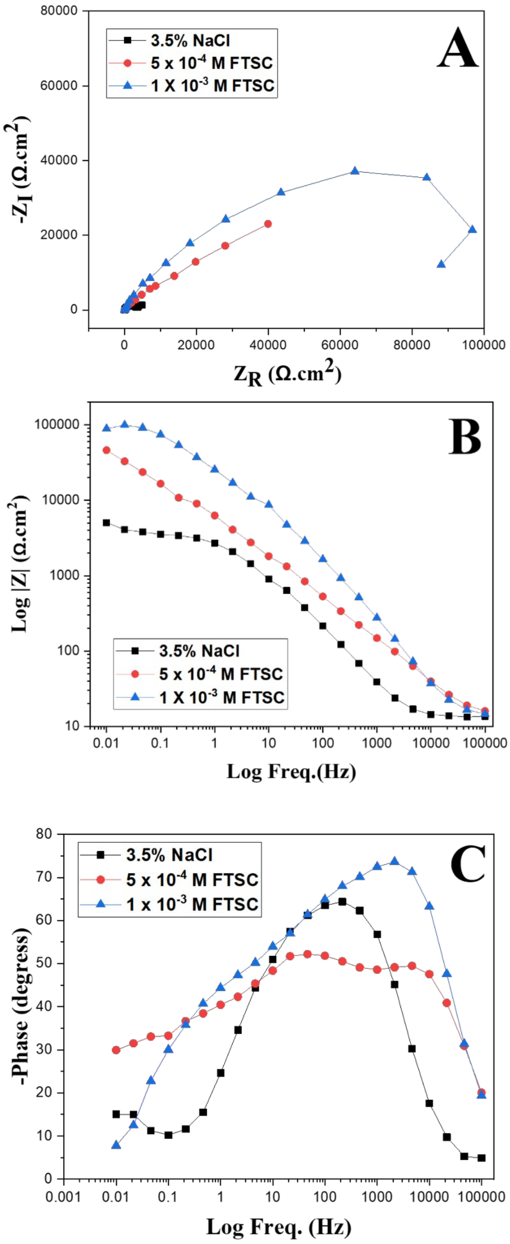

Figure 4: Electrochemical impedance spectroscopy (EIS) for brass in 3.5% NaCl without and with the presence of FTSC, Nyquist curve (A), and Bode curves (B-C).

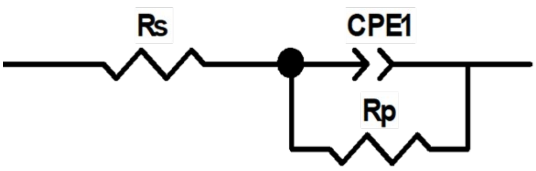

Figure 5: Equivalent Circuit for brass without inhibitor and with presence of inhibitor FTSC**.**

Adsorption isotherm

The most significant advance in the consumption restraint process is the adsorption of an inhibitor on the metal surface. Distinctive adsorption isotherms, including Langmuir, Frumkin, and Freundlich, are frequently used to portray the adsorption component of inhibitors.

In this work the Langmuir adsorption isotherm, exhibited by equation as shown below, was observed to be the most reasonable to fit.

$$\frac{C_{inh}}{\Theta}=\frac{1}{K_{ads}}+C_{inh} \qquad (4)$$

Where C_{inh} is the concentration of inhibitor, θ is the level of surface inclusion by inhibitor and K is the equilibrium constant of adsorption.

The plot of Cinh/θ versus inhibitor fixation (Cinh), (Figure 5) is straight, showing that the Langmuir adsorption isotherms material to portray the adsorption of FTSC. The estimation of the relationship coefficient (R^2) affirms that the adsorption of FTSC pursues the Langmuir isotherm. Additionally, this estimation of R^2 demonstrates that the compound atoms formed a monomolecular layer on the terminal surface (26). Further, the Gibbs free vitality of adsorption ($-\Delta G_{ads}$) is determined by equation as shown below:

$$\Delta G_{ads}=-RT\times\ln(55.5\times K_{ads}) \qquad \textbf{(5)}$$

The values of ΔG_{ads} around -20 kJ mol^{-1} imply complete electrostatic association (physisorption) while those with - 40 kJ mol^{-1} or more negative represents chemical interaction (27,28). So, the present investigation involves the estimations of which was found – 33.8 kJ. mol^{-1} for FTSC which recommended the ΔG_{ads} association of the inhibitor with the brass surface via mixed (chemisorption and physisorption) (10).

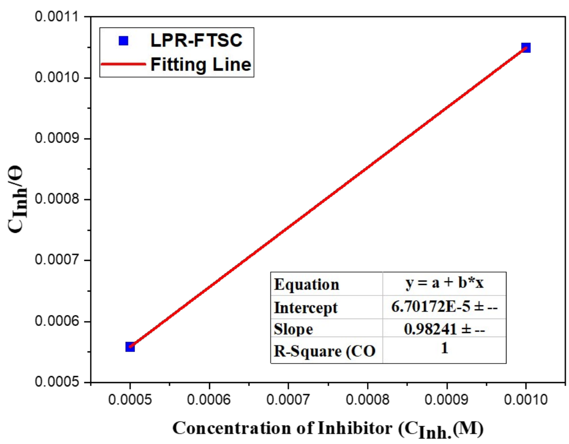

Figure 5: Langmuir adsorption isotherm of FTSC on the brass surface for 1 h.

Table 1: PDP and LPR for brass in 3.5% NaCl solution with and without the presence of FTSC.

Compound	*Concentration (M)*	*E_{corr} (V)*	*i_{corr} ($\mu A.cm^{-2}$)*	*$-\beta_c$ (V/dec)*	*β_a (V/dec)*	*θ*	*% IE*	*R_{lp} ($\Omega.cm^2$)*	*θ*	*% IE*
FTSC	3.5%NaCl	-0.221	16.5	0.641	0.165	-----	------	3058.7	-----	-----
	5×10^{-4}	-0.329	0.470	0.14	0.052	0.9715	97.51	29326.8	0.8957	89.57
	1×10^{-3}	-0.251	0.284	0.223	0.034	0.9828	98.28	64908.6	0.9529	95.29

Table 2: EIS for brass in 3.5% NaCl solution without and with the presence of FTSC

Sample	*Conc. (M)*	*R_S ($\Omega.cm^2$)*	*R_{p1} ($\Omega.cm^2$)*	*CPE_1 ($\mu F.cm^{-2}$)*	*n_1*	*R_{pT} ($\Omega.cm^2$)*	*% IE*
FTSC	Blank	5.94	2024.1	82.2	0.75	2024.1	------
	5×10^{-4}	3.86	43298	41.3	0.78	43298	95.32
	1×10^{-3}	4.33	45673	32.2	0.83	45673	95.60

4. CONCLUSION

The following conclusion may be drawn according to the collected data:

- The compound FTSC appeared to be an excellent corrosion inhibitor for brass in 3.5%NaCl solution and maximum efficiency reached 98.28% at 1×10^{-3} M and immersion time 1 h.
- Tafel curve showed that the corrosion potential shifted to a more negative value with the presence of FTSC and it can be classified as a cathodic type inhibitor.
- For EIS graphs with and without FTSC, there is a single equivalent circuit. The polarizing resistance rises when FTSC is present in a 3.5% NaCl solution, which lowers the double layer capacitance.
- A Langmuir adsorption model is the best match for the adsorption of FTSC molecules on the brass surface.

5. ACKNOWLEDGEMENT

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