



Experimental and Theoretical Evaluations: Green Synthesis of New Organic Compound bis ethanethiyl oxalamide as Corrosion Inhibitor for Copper in 3.5% NaCl



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Abstract

A newly synthesized (carbamothiol) oxalamide derivative N1, N2-bis (2-(((2-((2-aminoethyl) amino) ethyl) amino) ethanethiyl) oxalamide (**bis N**), was investigated for its ability to suppress corrosion of copper in 3.5% NaCl. The chemical structure was demonstrated using FT-IR, ¹H-NMR, and ¹³C-NMR. According to weight loss (WL), potentiodynamic polarization (PDF), and electrochemical impedance spectroscopy (EIS) techniques, the synthetic inhibitor exhibits higher corrosion inhibition efficiency. The corrosion rate decreases and the inhibition efficiency increases linearly with the concentration of inhibitor, reaching 93.3% at 0.01M. The adsorption of **bis N** obeys the Langmuir adsorption isotherm. The calculated adsorption isotherm parameter ΔG_{ads} is a negative value equals to -10.14 KJ/mol, suggesting that the **bis N** is adsorbed on the copper surface and achieves the spontaneous process. The efficiency of **bis N** for metal protection enhancement is assessed using density functional theory (DFT). Evaluation and discussion of quantum different descriptors are also included.

Keywords: Copper corrosion; inhibition; potentiodynamic polarization; electrochemical impedance; sodium chloride; DFT.

I. Introduction

The fifth-most common metal on the planet, copper, is very beneficial both in its pure form and as an alloy [1]. Because of its high thermal and electrical conductivity and mechanical usefulness, Cu is also used in various items, including conductors, heat exchangers, and the electronic industry [2,3]. Copper has a relatively noble potential, despite this, it corrodes far more quickly in chloride and seawater conditions [4,5]. It is widely known that the amount of chloride ions in the environment affects how easily copper dissolves anodically. The formed CuCl, which is not sufficiently protective, and dissolves when it reacts with excess chloride [6–8]. The inhibitors can be introduced to the medium or coated on the metal

surface to slow down the corrosion process to avoid corrosion in the metal used in such corrosive media, both methods work well [9–13].

Several investigators have reported [5–8] that organic compounds can act as possible corrosion inhibitors for numerous metals and alloys in various corrosive media.

Heterocyclic compounds having N, O, S, P, or p-systems make up many effective corrosion inhibitors by generating a physical barrier to reduce the transport of corrosive species to the metal surface [14,16].

Organic compounds that include conjugated double bonds and hetero atoms especially those, that can form polymeric complexes with copper by coordinating with Cu⁰, Cu⁺, or Cu²⁺ through their lone pair electrons [17]. These adhere to the copper

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surface to produce a protective coating that prevents corrosion by serving as a wall against harmful ions like chloride [18].

The synthesized derivative of the carbamothioyl moiety is favored over that of the regular urea moiety in moderate circumstances, because the thiourea derivatives' preparation was done in harsh conditions [19,20]. As well as the high activity of thiourea as anticorrosion compound [20-22], for alloys and metals, amid compounds are frequently employed as corrosion inhibitors. A protective coating is created by utilizing a tiny inhibitor dose to delay the corrosion reaction [23,24].

Using inhibitors is the main method for metal and alloy protection. The green inhibitors are currently promising due to minimizing environmental impact and offering efficient inhibition. The organo- sulphur and heterocyclic compounds derivative have the basic requirements to be considered eco-friendly formulations. These compounds are adsorbed on the metal surface by blocking the active sites during the inhibition process. This leads to the need of development of new environmentally-friendly inhibitors for copper metal.

A newly synthesized (carbamothiol) oxalamide derivative N1, N2-bis (2-(((2-aminoethyl) amino) ethyl) amino) ethanethioyl) oxalamide (**bis N**), was investigated for its ability to suppress corrosion of copper in 3.5% NaCl.

The evaluation of the corrosion behavior of copper in aggressive media in the absence and presence of newly synthesized **bis N** using potentiodynamic polarization (PDP), impedance spectroscopy (EIS), and weight loss methods were discussed. The use of FT-IR, ¹H-NMR, and ¹³C-NMR validated the structure.

2. Experimental

2.1 Instrumentation

The spectra of FTIR of the synthesized sample were recorded using Pye Unicam SP3-300 spectrophotometer. The technique of the KBr disks is applied. Also, the measurements of both ¹H-NMR and ¹³C-NMR are detected by Bruker WP AC 300(300 MHz). DMSO-d₆ is used as solvent and the internal standard is tetramethylsilane (TMS). The chemical shifts (δ) are detected ppm). Elemental analyses are also carried out.

2.2. Preparation of bisN:

Oxalyl dichloride (0.01 mol, 1.26 g) and ammonium thiocyanate (0.02mol, 1.52g) were mixed and stirred in dry acetone for 20 min. Then, ammonium chloride was removed from the product by filtration to collect oxalyl diisothiocyanate (1). The filtrate is added during stirring to 0.02 mol ethane-1, 2-diamine (2),

for 15 min. Then, the acetone evaporates, and the precipitate is washed using petroleum ether 40-60 several times, affording compound namely bis N (N₁, N₂-bis(2-((2-((2-aminoethyl)amino) ethyl) amino) oxalamide a dark brown oily product, Yield 92%, [25].

IR (KBr, ν /cm⁻¹): 3615, 3540cm⁻¹ (ν_{NH / NH₂}); 1730 cm⁻¹ (ν_{C=O}), ¹HNMR(300 MHz, DMSO-d₆): δ(ppm): 1.2(s, 4H.2 NH₂, D₂O exchangeable), 1.8 (trip, 4H, 2 CH₂ next to NH₂), 3.1(trip, 4H, 2CH₂ next to NH), 7.1 (s .2H, 2NH D₂O exchangeable next to C=S), 9.1 (s .2H, 2NH D₂O exchangeable next to C=O), ¹³C NMR(DMSO-d₆,) δ(ppm): 50.53, 50.73 (2CH₂-NH), , 85.53 (2CH₂-NH₂), 168.38 (2C=O), 187.59 (2C=S). Calculated for C₈H₁₆N₆O₂S₂ (292.38): C, 32.86; H, 5.52; N, 28.74; S, 21.93 Found: C, 32.50; H, 5.35; N, 28.40; S, 21.80 [26].

2.3. Corrosion testing

3.5% NaCl was used as an aggressive solution. The synthesized chemical bis N ranged from 10⁻³ to 10⁻¹ M used as an inhibitor. Copper 1x 1x 3 cm³ was used as a working electrode, polished to a maximum of 1200, washed with ethanol, and then dried.

2.3.1. Weight loss

After washing, the copper specimens were immersed in 3.5% NaCl with and without bis N for three hours. Copper was weighed before and after immersion. The average weight loss of three samples was determined. Equations (1) used to determine the corrosion rate (CR) and inhibition efficiency (IE %) [27]:

$$\text{Corrosion rate } (C_R) = \frac{(W - W_0)}{AT} \quad (1)$$

Where, W and W₀: the weight before and after immersion, A: surface area, and T: immersion time in h.

$$IE\% = \frac{C_R - C_{R(i)}}{C_R} \times 100 \quad (2)$$

Where, C_R and C_{R(i)} are the corrosion rate of the copper in the absence and presence of different concentrations of **bis N**.

2.3.2. Potentiodynamic polarization (PDP)

The PDP measurements were performed using a computerized (Potentiostat/ Galvanostat Autolab PG STAT 30). A three-electrode cell was used, with Pt as a counter electrode, a saturated calomel electrode (SCE) as a reference electrode, and a copper sample as the working electrode. Starting at -0.3 V below steady-state potential, the scan rate of 1 mV s⁻¹. The polarization curves were used to determine the corrosion parameters corrosion potential (E_{corr}), corrosion current (I_{corr}), and corrosion rate (CR) using Autolab software.

$$IE\% = \frac{I_0 - I_1}{I_0} \times 100 \quad (3)$$

Where: I₀ and I₁ are the densities of the corrosion currents without and with bis N, respectively. The following expression can be used to compute the degree of surface coverage (θ):

$$\theta = \frac{C_R - C_{R(i)}}{C_R} \quad (4)$$

2.3.3. Electrochemical impedance spectroscopy (EIS)

EIS measurements using a voltage signal of 10 mV amplitude were performed at open circuit potential and frequencies ranging from 0.1 Hz to 100 KHz. The Auto Lab program was used to analyze the EIS curves and predicts the impedance parameters: charge transfer (R_{ct}), solution resistance (R_s), and double-layer capacitance (C_{dl}). The percentage inhibition efficiency was calculated using the following equation:

$$IE\% = \frac{R_1 - R_0}{R_1} \times 100 \quad (5)$$

Where, R_1 and R_0 : the charge transfer resistance with and without the inhibited solutions.

2.4. Theoretical calculation

The quantum chemical computations of the synthesized compound were carried out in this study utilizing the density functional theory (DFT) contained in the Hyper Chem program. The B3LYP program function was used to optimize the geometry. The energy of the highest and the energy of the lowest empty molecular orbital (E_{HOMO}) and (E_{LUMO}), respectively was used to evaluate relevant descriptors such as energy gap and electronegativity.

1. Results and Discussion

3.1. Characterization

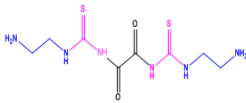
Oxalyl diisothiocyanate (1) is formed by the reaction of oxalyl dichloride with ammonium thiocyanate. Followed to the filtrate is added to amines (2) to prepare thiourea derivative (**bis N**) (see **Scheme 1**).

Fig. 1 shows the IR spectra of the prepared compound **bis N** confirming the function groups of the structure. IR spectrum of the **bis N** confirms the presence of carbonyl, acetyl and the amino groups at 1730 cm^{-1} and $3615, 3540\text{ cm}^{-1}$, respectively [28].

3.2. Weight loss (WL)

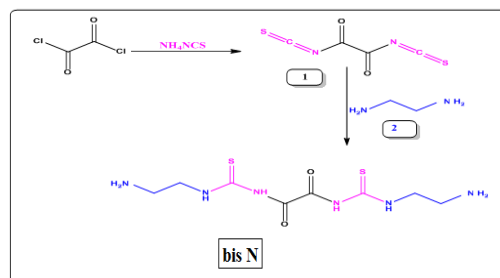
Fig. 2 shows a change in the weight loss of copper in 3.5% NaCl with and without the synthetic **bis N** compound. The degree of the loss in weight decreased as the concentrations increased. By using

Table 1: Structure, IUPAC name, and chemical formula of bisN

| Comp. | Structural | IUPAC Name | Chemical Formulas | Active center |
|--------------|---|---|--------------------|---------------|
| Bis N |  | $N_{1,2}$ -bis(2-((2-aminoethyl)amino)ethyl)amino ethanethioyal oxalamide | $C_8H_{16}N_6O_2S$ | |

equ.1, 2 the inhibition efficiency (IE) and corrosion rate (CR) were estimated and listed in **Table 2**.

$^1\text{H-NMR}$ spectra showed multiplet band at 3.1 – 1.8 ppm and a singlet at 7.1 ppm assigned to imino groups. In addition, the $^{13}\text{C-NMR}$ spectra for **bis N** exhibited the presence of a 53, 50.73 ($2\text{CH}_2\text{-NH}$), 85.53 ($2\text{CH}_2\text{-NH}_2$). Also, signal related to carbonyl group assigned at δ 168.38 ppm. The C=S is located at δ 187.59 ppm.



Scheme 1 The formation of thiourea derivative (**bis N**)

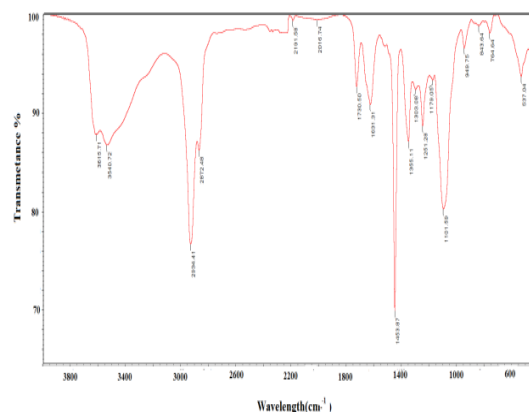


Fig. 1 FT-IR spectra of the **bis N**.

$^1\text{H-NMR}$ (300 MHz, $\text{DMSO-}d_6$): δ (ppm): 1.2(s, 4H, 2 NH_2 , D_2O exchangeable), 1.8 (trip, 4H, 2 CH_2 next to NH_2), 3.1(trip, 4H, 2 CH_2 next to NH), 7.1 (s .2H, 2NH D_2O exchangeable next to C=S), 9.1 (s .2H, 2NH D_2O exchangeable next to C=O).

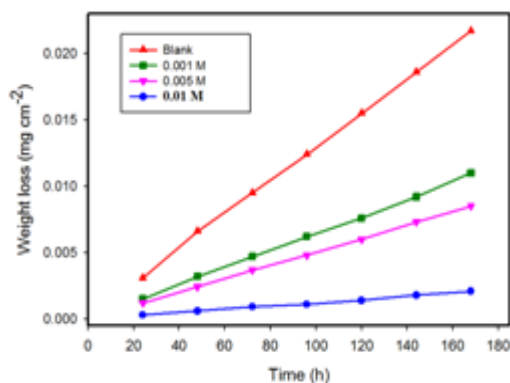


Fig. 2 Weight loss- time curves for copper in 3.5% NaCl in the absence and presence of different concentrations of synthetic compound **bis N** at 25°C.

CR is significantly lower in the presence of different concentrations of **bis N** than in the blank solution as seen in Fig 3. This result demonstrated the suppression of copper corrosion in a 3.5% NaCl solution due to the development of a barrier film that prevents the chloride from reaching the copper surface. The inhibition efficiency at 0.01 M is 90.32% due to the active center's existence as amino, thio, and carbonyl groups [29,30].

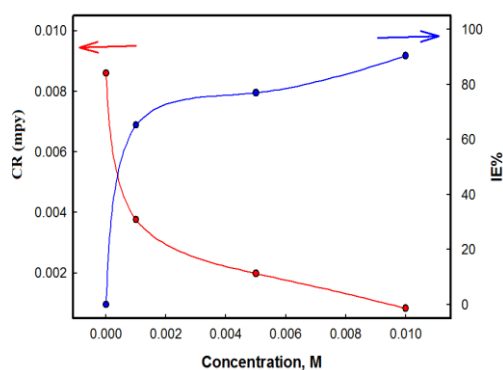


Fig.3 The variation of CR and IE% of copper with **bis N** concentration in 3.5M NaCl solution obtained by weight loss method at 25°C.

Table 2 Corrosion rate (CR), surface coverage (θ) and the inhibition efficiency (IE%) of copper metal in 3.5% NaCl solution in the absence and presence of different concentrations of **bis N** inhibitor obtained by weight loss method at 25°C.

| Inhibitor | Concentration (M) | (CR) (mpy) | (θ) | (IE %) |
|--------------|-------------------|------------|--------------|--------|
| Blank | 0 | 0.0086 | - | - |
| Bis N | 0.001 | 0.00337 | 0.608 | 60.82 |
| | 0.005 | 0.00174 | 0.797 | 79.72 |
| | 0.01 | 0.00083 | 0.903 | 90.32 |

3.3. Electrochemical impedance spectroscopy (EIS)

The EIS is a non-destructive technique that offers several characteristics. Fig. 4a shows the Nyquist plot for copper in 3.5% NaCl with and without the

bis N inhibitor. The findings demonstrate the semicircle in all concentrations and show that its diameter grows as concentration increases, indicating that the charge transfer mechanism is the primary cause of corrosion [31].

Fig.4 (b, and c) show the variation of the $\log |Z|$ and phase angle with frequency. The impedance amplitude increases with concentration reaching a log maximum of 3.3 ohm cm^2 at 0.01 M compared to 2.0 ohm cm^2 in the absence of the inhibitor. This is due to the presence of two amino groups. The equivalent circuit used to model the experimental data is shown in Fig.4d. Constant phase element CPE was used instead of capacitance C for better fitting [32].

The values of the EIS from fitting parameters such as R_s , R_{ct} , C_{dl} , and IE are listed in Table 3. As seen from the table, R_{ct} values increased linearly with the concentration of the inhibitor reaching a maximum value of 12.645 $\text{k}\Omega \cdot \text{cm}^2$ while CPE values decreased to 4.31×10^{-5} F at 0.01 M of **bis N**. The creation of an adsorbed film of the **bis N** inhibitor molecules mostly is the cause of the increased R_{ct} and decreased double-layer capacitance. The increase in inhibition efficiency with the increase in inhibitor concentration is attributed to the **bis N** inhibitor's ability to cover more surface area by adsorption. The maximum inhibition efficiency calculated from EIS data is 93.3% for copper with 0.01M **bis N**. This confirms that the **bis N** can act as an effective inhibitor for copper in 3.5% NaCl.

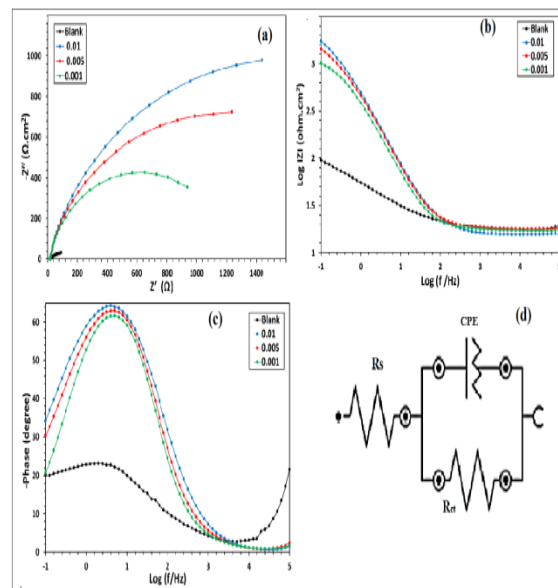


Fig. 4 (a) Nyquist, (b) Bode, and (c) phase plots and (d) equivalent circuit of copper in 3.5% NaCl with different concentration of **bis N**.

Table 3 The values of the EIS parameters for investigated compound **bis N**

| Compound ds | Conc. (M) | E_{corr} (V) | β_a (V dec ⁻¹) | β_c (Vdec ⁻¹) | I_{corr} (Acm ⁻²) | CR (mpy) | IE (%) |
|-------------|-----------|----------------|----------------------------------|---------------------------------|---------------------------------|----------------------|--------|
| Blank | 0 | -0.087 | 2.34×10^{-2} | 1.09×10^{-2} | 1.19×10^{-5} | 2.8×10^{-1} | ----- |
| Bis N | 0.001 | -0.211 | 2.42×10^{-2} | 4.55×10^{-2} | 6.66×10^{-6} | 1.5×10^{-1} | 44.6 |
| | 0.005 | -0.216 | 1.73×10^{-2} | 1.76×10^{-2} | 2.07×10^{-6} | 4.8×10^{-2} | 82.8 |
| | 0.01 | -0.216 | 1.88×10^{-2} | 3.99×10^{-2} | 2.88×10^{-6} | 3.4×10^{-2} | 87.9 |

3.4. Potentiodynamic polarization measurements

Fig. 5 displays the PDP of copper with and without various **bis N** concentrations in 3.5% NaCl solution. The curves demonstrated that the addition of compound **bis N** affects both the cathodic and anodic performance, indicating that **bis N** can reduce copper anodic dissolution. It is seen from the figure that the current density (I_{corr}) decreases with **bis N** concentrations indicating the creation of a protecting film on the copper surface improved the surface coverage [33].

It is clear from Fig. 5 that the anodic branch exhibits three distinct regions: first, the dissolution of copper metal to Cu^+ increases current density; second, cuprous chloride formation causes a decrease in current density; and third, a sudden increase in current causes copper to dissolve and form cupric chloride.

Table 4 shows the electrochemical characteristics, including current density (I_{corr}) and anodic and cathodic slopes (β_a , β_c), that were determined by extrapolation at the corrosion potential (E_{corr}). Equation 3 was used to evaluate IE%. According to

Table 4 The values of the polarization parameters for investigated compound **bis N**

| sample | Conc. (M) | R_s (Ω) | R_{ct} ($K\Omega\text{ cm}^2$) | CPE (μmho) | n | IE % |
|--------|-----------|--------------------|------------------------------------|-------------------------|-------|------|
| Blank | 0 | 17.17 | 0.139 | 1.01×10^{-2} | 0.500 | ---- |
| Bis N | 0.001 | 2.2 | 1.1 | 4.01×10^{-4} | 0.655 | 85.1 |
| | 0.005 | 1.83 | 1.247 | 3.35×10^{-4} | 0.572 | 88.8 |
| | 0.01 | 14.03 | 2.097 | 7.34×10^{-5} | 0.623 | 93.3 |

3.5. Adsorption isotherm

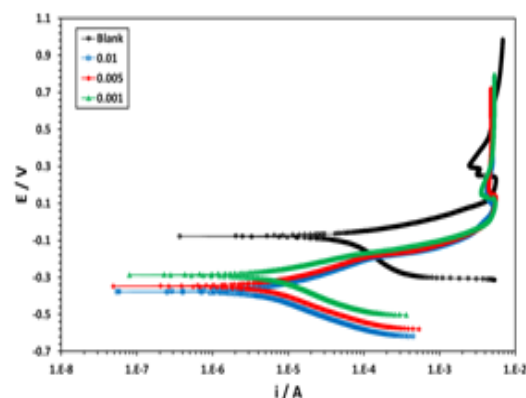
A protective coating can be created by the organic compound **bis N** interacting with the copper surface through adsorption. The produced (carbamothioyl) oxalamide derivative molecules' addition of nitrogen, sulfur, and oxygen enhances the adsorption process [1].

The Langmuir isotherm was found to graphically fit the experimental data [34,35]:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad (7)$$

Where C_{inh} is the concentration of **bis N**, the degrees of the surface coverage (θ), and K_{ads} is the adsorption equilibrium constant. The plot of C_{inh}/θ vs C_{inh} as shown in Fig 6 gave straight lines obeyed Langmuir isotherm with 0.995 correlation coefficient R value

polarization measurements, the inhibition efficiency increases with **bisN** concentration, due to the increasing of the blocked fraction of the copper surface by adsorption. The inhibition efficiency reached 87.9% inhibition efficiency at 0.01 M providing more effective inhibition than other doses.

**Fig. 5** Anodic and cathodic polarization curves in absence and presence of different concentration of **bis N** in 3.5 % NaCl solution.

[36], slope equal 1.0, and intercept give the value of K_{ads} . The adsorption equilibrium constant used to find out the thermodynamic function as the standard free energy ΔG_{ads} according to: [1]

$$\Delta G_{ads} = -RT \ln(55.5K_{ads}) \quad (8)$$

Where T: temperature (K), R gas constant and 55.5 is the concentration of H_2O molecules

The calculated ΔG_{ads} is a negative value ($-13.88\text{ KJ mol}^{-1}$), suggesting that the **bis N** is adsorbed on the copper surface and achieves the spontaneous process. This value suggests that the adsorption is physisorption and involves electrostatic interaction [32,37].

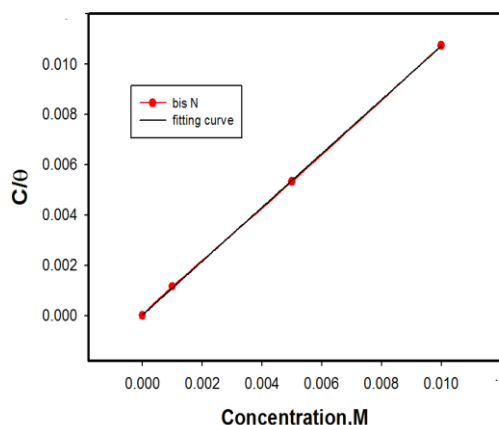


Fig 6 The Langmuir adsorption isotherm of copper in 3.5% NaCl in the presence of compound **bis N** at 25 °C.

3.6. Computational Studies

Table 5 provides a summary of the calculated quantum chemical characteristics, including (E_{HOMO}), (E_{LUMO}), energy gap ($E_{\text{g}} = E_{\text{LUMO}} - E_{\text{HOMO}}$), global hardness η and energy of back donation ($E_{\text{b-d}} = \eta/4$) [38]. **Fig. 7** demonstrates that the HOMO and LUMO energy orbitals were localized on the Sulfur, Nitrogen of the molecule's center and almost nonexistent on the terminal hetero atom, indicating that the synthesized molecule has good adsorption centers and supporting the theory of several studies that the hetero atoms and p electrons are both the controlling factor for the inhibition activity [39].

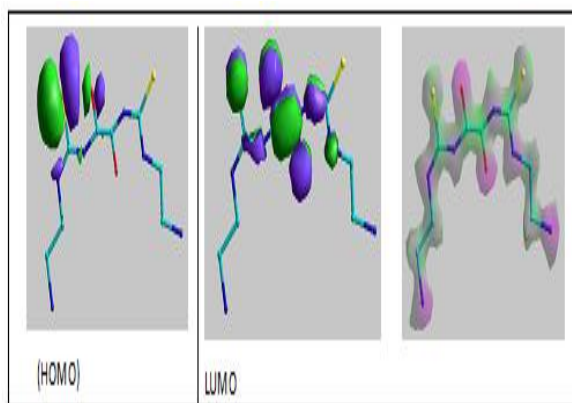


Figure 7: HOMO, LUMO and the electrostatic potential, magenta indicates a most electron rich region, lime green refers to the positive region.

It is widely believed that the bigger the value of E_{HOMO} (-7.68 eV), the easier it is for an inhibitor to donate electrons to an unoccupied d orbital of a copper atom, and the greater the effectiveness of the inhibition. The easier it is for copper atoms to absorb electrons to form feedback bonds, the lower the energy of unoccupied molecular orbitals must be ($E_{\text{LUMO}} = -0.34$). Another crucial factor that must be

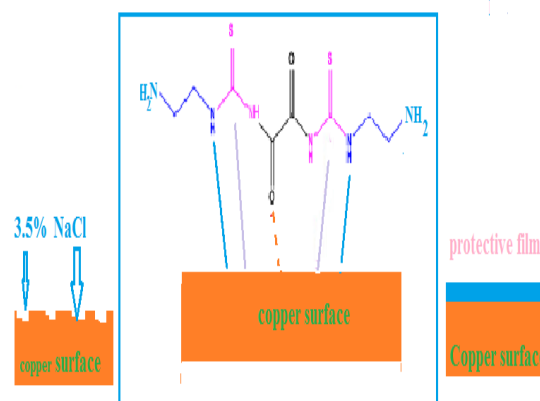
taken into account is the energy gap. Higher inhibition efficiency is achieved by an inhibitor with a smaller value of $\Delta E_{\text{g}} = 7.34$ eV. In addition, the hardness of a molecule is used to determine its reactivity and was found to be 3.67. As the hardness of the inhibitors rises and the reactivity of the molecule decreases and increases the deformation resistance of the electrons cloud of atoms [24]. The determined values of $E_{\text{b-d}}$ are related to the global hardness, which was found to be 0.05 eV implying that the best inhibitor, back-donation is preferred. The previous data matches also the partial charge distribution around the molecule, which provides information on the molecule's reactive site.

Table 5: Quantum descriptor for bis N.

| Molecules | E_{HOMO} | E_{LUMO} | ΔE_{g} | η | $\Delta E_{\text{d-b}}$ |
|--------------|-------------------|-------------------|-----------------------|--------|-------------------------|
| Bis N | -7.68 | -0.34 | 7.34 | 3.67 | 0.05 |

3.7. Mechanism of corrosion inhibition

For copper surfaces in 3.5% NaCl solution, the inhibition of **bis N** depends on the adsorption that creates a barrier. The two crucial reactions of oxidation and reduction are what cause corrosion. Most likely, the organic molecules stop both reactions, preventing corrosion. The inhibition mechanism of **bis N** can be considered as physical adsorption from Langmuir isotherm. The adsorption takes place by the interaction of unpaired electrons of nitrogen, oxygen, and sulfur of amino, carbonyl, and thio-groups with the copper surface. To confirm the presence of the adsorbed film on the copper surface, several techniques were used such as WL, PDP, and EIS. Corrosion of the copper was attacked by an aggressive solution without inhibitor, and the adsorption of **bis N** molecules which form a protective layer at a high concentration of **bis N** are presented in **Scheme 2**.



Scheme 2 Schematic representation for adsorption process of **bis N** on copper surface in 3.5% NaCl

Conclusions

New Green synthesis of new organic compound N1, N2-bis (2-(((2-((2-aminoethyl) amino) ethyl) amino) ethanethioly) oxalamide (**bis N**) was prepared and characterized by elemental, IR, ¹H-NMR and ¹³C-NMR. The inhibitor's efficiency was evaluated gravimetrically and electrochemically (PDF, EIS).

In light of the information, the following conclusions are made:

- **bis N** is a good corrosion inhibitor for copper in 3.5% NaCl solution.
- The inhibition efficiency of **bis N** rose with the concentration, reaching a maximum of 93.3%.
- The inhibiting efficiencies obtained by all measurements are in good agreement.
- bisN adsorption follows the Langmuir isotherm
- The negative sign of the ΔG_{ads} indicates a spontaneous mechanism for the adsorption of **bisN** on the copper surface.
- The theoretical and experimental data are well correlated in the quantum chemical computations utilizing DFT.

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Conflict of interest:

No conflict exists and the authors declare that they have no conflict of interest.

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