ANADOLU ÜNİVERSİTESİ BİLİM VE TEKNOLOJİ DERGİSİ – A Uygulamalı Bilimler ve Mühendislik

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CORROSION INHIBITION OF BENZOTRIAZOLE AND *N*-MESYL BENZOTRIAZOLE ON LOW CARBON STEEL IN SULFURIC ACID

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ABSTRACT

This study examines usage of benzotriazole (Bt) and *N*-mesyl benzotriazole (BtSO₂CH₃) for improvement of corrosion resistance of low carbon steel (LCS) in 0.1 M sulfuric acid (H₂SO₄). The inhibitor effect of Bt and BtSO₂CH₃ on the corrosion behavior of LCS was studied using electrochemical impedance spectroscopy (EIS), potentiodynamic polarization curves and polarization resistance techniques. Under investigated experimental conditions these compounds showed a good corrosion inhibition. The inhibition efficiency of BtSO₂CH₃ was more than that of Bt. The potentiodynamic polarization curves indicated that compounds are mixed type inhibitors. The inhibiting properties of compounds were found to depend on the concentration. The adsorption mechanism of compounds was investigated using different adsorption isotherms. The equilibrium constants and free energies of adsorption using the adsorption isotherms were calculated.

Keywords: Benzotriazole, Corrosion, Inhibitor, Carbon steel.

SÜLFÜRİK ASİT İÇERİSİNDE DÜŞÜK KARBON ÇELİĞİNE BENZOTRİAZOL VE *N*-MESİL BENZOTRİAZOL'ÜN KOROZYON İNHİBİSYONU

ÖΖ

Bu çalışma 0,1 M sülfürik asit içinde düşük karbon çeliğinin (LCS) korozyon direncini arttırmak için benzotriazol (Bt) ve yeni sentezlenen *N*-mesil benzotriazol (BtSO₂CH₃) kullanımını incelemektedir. LCS'nin korozyon davranışına karşı Bt ve BtSO₂CH₃'ün inhibitör etkisi, elektrokimyasal empedans spektroskopisi (EIS), potansiyodinamik polarizasyon eğrileri ve polarizasyon direnci teknikleri kullanılarak çalışılmıştır. Belirtilen deneysel koşullarda bu bileşikler iyi bir korozyon inhibisyonu göstermektedir. BtSO₂CH₃'ün inhibitör etkinliği, Bt'ün inhibitör etkinliğinden daha fazladır. Potansiyodinamik polarizasyon eğrileri bileşiklerin karma inhibitör olduğunu göstermektedir. Bileşiklerin inhibisyon özelliklerinin derişime bağlı olduğu bulunmuştur. Bileşiklerin adsorpsiyon mekanizmaları farklı adsorpsiyon izotermleri kullanılarak incelenmiştir. Aynı zamanda serbest enerji ve denge sabiti değerleri adsorpsiyon izotermlerinden hesaplanmıştır.

Anahtar Kelimeler: Benzotriazol, Korozyon, İnhibitör, Karbon çeliği.

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1. INTRODUCTION

The corrosion of metals occurs in higher rates in acidic media especially when soluble corrosion products are formed. Sulfuric acid is an industrial acid used in many processes including etching, pickling anodizing, de-scaling, acid cleaning, oil-well acidizing etc., and damages the materials under treatment (Abdel-Gaber et al. 2006). The use of inhibitors is one of the most practical methods for protection against corrosion, especially in acidic media (Trabanelli 1991).

The most important acid inhibitors are organic compounds containing heteroatom(s) like nitrogen, sulfur and oxygen. The organic compounds containing nitrogen atoms are preferred in HCl media, on the other hand organic compounds containing sulfur atoms are preferred in H₂SO₄ media (Abd El-Maksoud and Fouda 2005; Bentiss et al. 2000; Abd El-Maksoud 2003). Also corrosion inhibition effects of organic compounds containing both nitrogen and sulfur atoms have been studied by several works (Bentiss et al. 1999: Kertit and Hammouti 1996; El-Shafei et al. 2001). The effect of these compounds as inhibitor can be described as altering the nature of the corrosion product, changing the type of adsorption on the metal surface and rearranging the corrosion potential and current (Gomma 1998a).

In the present paper, the corrosion behavior of low carbon steel in 0.1 M sulfuric acid was investigated using electrochemical methods, namely electrochemical impedance spectroscopy (EIS), potentiodynamic polarization curves (Tafel plots) and polarization resistance technique. The inhibitor effect of benzotriazole (Bt) and newly synthesized *N*-mesyl benzotriazole (BtSO₂CH₃) was examined in H_2SO_4 media. The adsorption behavior of the compounds was tested in order to determine the best fitted adsorption isotherm and thermodynamic parameters.

2. MATERIALS AND METHODS

2.1 Corrosion Tests of Benzotriazole and *N*-Mesil Benzotriazole

N-Mesil benzotriazole was synthesized using literature procedure (Katritzky et al. 1992) and benzotriazole was obtained from Aldrich (Table 1).

Anadolu Üniversitesi Bilim ve Teknoloji Dergisi - A 12 (1) Uygulamalı Bilimler ve Mühendislik

Corrosion tests were carried out in a three electrode type cell using the reference electrode (Ag/AgCl) in a luggin capillary and the counter electrode (Pt wire). The working electrode was prepared from a cylindrical low carbon steel rod (0.3% Mn, 0.1% C, 0.05% S, 0.04% P and remaining is iron) insulated with polyester resin such that the area exposed to solution was 0.1548 cm². Distilled water and analytical reagent grade sulfuric acid was used for solutions. The electrode was polished with 600, 800, 1000 and 1200-grit emery papers prior to each experiment using Metkon Grinding and Polishing Machine. Prior to measurements, working electrode was pre-treated with ethyl alcohol and acetone solution to remove the impurities of the metal in an ultrasonic cleaner. Electrochemical corrosion measurements were investigated using electrochemical impedance spectroscopy (EIS), potentiodynamic techniques. The impedance spectra were recorded and *potentiodynamic* measurements were carried out after 1 h and 2 h of exposure of the electrode in the test solution, respectively. Electrochemical impedance measurements were obtained at measured E_{ocp} values applying 8 mV of amplitude, in frequency range from 10⁵ to 10⁻² Hz. *Potentiodynamic* polarization studies was performed with a scan rate of 2 mV/s in the potential range from -400 to +400mV relative to the corrosion potential. All corrosion measurements were performed in 0.1 M H₂SO₄ solution at room temperature and open air with Gamry Model Reference 600 Potentiostat/Galvanostat. The analysis of the impedance spectra and fitting of the experimental results to equivalent circuits was performed using ZSimpWin 3.21. Each experiment was repeated four times to check the reproducibility.

3. RESULTS AND DISCUSSION

3.1 Corrosion Tests

3.1.1 Potentiodynamic Polarization

Figure 1 shows cathodic and anodic potentiodynamic polarization curves of LCS in 0.1 M H_2SO_4 without and with various concentrations of compounds Bt and BtSO₂CH₃.

It is observed that the addition of Bt and $BtSO_2CH_3$ affects cathodic and anodic branch of potantiodynamic polarization curves of LCS. In this case both compounds act as mixed type inhibitor. Also the small shift of the corrosion potential in the positive sense as compared to the uninhibited solutions shows that the effect on the anodic reaction is somewhat more pronounced than that on the cathodic reaction (Larabi et al. 2006).

| Structure of molecules | Name of molecules | Abbreviation |
|------------------------------------|-----------------------|-----------------------------------|
| N, N, N, N, H | Benzotriazole | Bt |
| N N O=S=O CH ₃ | N-mesyl benzotriazole | BtSO ₂ CH ₃ |

Table 1. The investigated molecules for inhibition efficiency

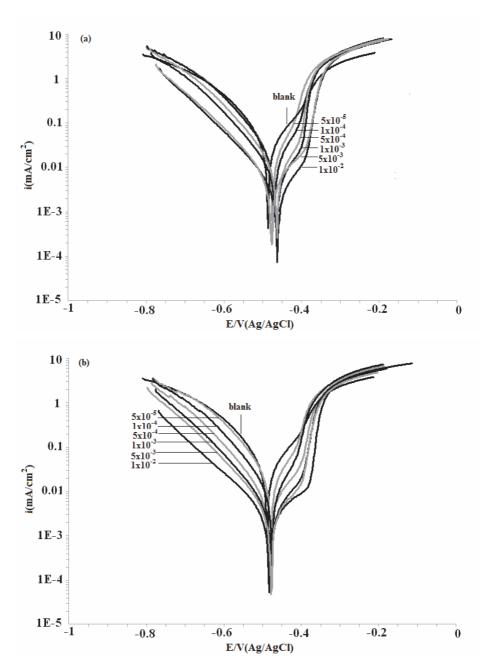


Figure 1. Polarization curves of LCS obtained in $0.1 \text{ M H}_2\text{SO}_4$ in the presence of various concentration of (a) Bt (b) BtSO₂CH₃.

4

The corrosion current densities (i_{corr}) and corrosion potentials were calculated from the intersection of Tafel lines extrapolation. Moreover, cathodic and anodic Tafel constants (β_c and β_a) were determined in all cases directly from the slope of Tafel plots. The calculated parameters were given in Table 2.

This table shows that corrosion current density decreased with increasing concentration of inhibitors. The percentage inhibition efficiency at different inhibitor concentrations was calculated using the following equation:

$$IE(\%) = \left[1 - \frac{i_{corr\ (inh)}}{i_{corr\ (blank)}}\right] \times 100$$

where, $i_{corr}(inh)$ and $i_{corr}(blank)$ are the corrosion current densities for in the presence and absence of inhibitor, respectively. Inspections of the percentage inhibition efficiency show that inhibition efficiency increases with an increase in the inhibitor concentration. This result indicated that Bt and BtSO₂CH₃ are adsorbed at the steel surface.

3.1.2 Electrochemical Impedance Spectroscopy

Nyquist plots of LCS in the 0.1 M H_2SO_4 at room temperature in the presence and absence of Bt and BtSO₂CH₃ are given in Figure 2. Curves in plots show almost semicircles. It can be seen from Figure 2 that the Nyquist plots consist of one capacitive loop in the high frequency. The high frequency loop was attributed to the charge transfer resistance and double capacitance (Larabi et al. 2006).

The capacitive loop is indicated to the relaxation of the double layer since its chargingdischarging is very rapid process (Gasparac et al. 2000; Diard et al. 1998). The impedance spectra can be analyzed with the electrical circuit in Figure 3. It consists of solution resistance, R_s , a constant phase element, Q, in parallel with polarization resistance which contains charge transfer resistance and adsorbed molecules resistance, R_p . The constant phase element can be attributed to the relaxation of the double layer which means the time required for returning of the charge distribution to equilibrium after an electric perturbation (Popova and Christov 2006).

The impedance parameters derived from the Nyquist plot and the percent protection effi-

Anadolu Üniversitesi Bilim ve Teknoloji Dergisi - A 12 (1) Uygulamalı Bilimler ve Mühendislik

ciency values were also calculated by using the following equation;

$$IE(\%) = \left[1 - \frac{R_p(blank)}{R_p(inh)}\right] \times 100$$

where $R_p(inh)$ and $R_p(blank)$ are the polarization resistance with and without inhibitor, respectively. Table 3 shows calculated parameters. As can be seen from this table, R_p values of increase with the addition of Bt and BtSO₂CH₃ when compared with that without inhibitor. Furthermore the values of R_p increased with the increasing Bt and BtSO₂CH₃ concentration. This result can be related increasing surface coverage by the inhibitors leading to an increase in IE(%).

Although the R_p values increased with the increasing corrosion inhibitor concentration, the values of Q decreased with increase in the concentrations of the inhibitors. Q is constant phase elements (CPE) (Sherif and Park 2006). A constant phase element composed of component Q_{dl} and a coefficient n. The n quantifies different physical phenomena like surface inhomogeneity resulting from surface roughness, inhibitor adsorption, porous layer formation (Li et al. 2010). The double layer capacitance (C_{dl}) value is affected by imperfections of the surface and that this effect is simulated by imperfection of the surface. This effect is simulated via CPE. The double layer capacitance is deduced from the following equation (Qu et al. 2007).

$$C_{dl} = Q_{dl} \times (2\pi f_{max})^{n-1}$$

where f_{max} represents the frequency of which imaginary value reaches a maximum on the Nyquist plot. The value of *n* is used to account for the roughness of the electrode. The C_{dl} values decrease with increasing of inhibitor concentration. The decrease in C_{dl} values, which is generally result from a decrease in the dielectric constant and/or an increase in the double layer thickness, evolves out of inhibitor adsorption onto the metal/electrolyte interface (Oguzie et al. 2007). Parameter *n* increases in the inhibited solution compared to the pure acid. This is connected with a decrease in heterogeneity resulting from inhibitor adsorption. Our results are in agreement this phenomenon (Table 3).

Our results are more satisfactory than some literature results according to calculated inhibitor efficiency. In the literature following results were obtained for inhibition effect of Bt.

| Compounds | Inhibitor Con- | E_{corr} | <i>i</i> corr | IE | θ |
|-----------------------------------|--------------------|---------------|-------------------------|------|-------|
| | centration (M) | vs Ag/AgCl(V) | (Acm ⁻²) | (%) | |
| Blank | - | -0.487 | 243 x 10 ⁻⁶ | - | - |
| Bt | 5×10^{-5} | -0.475 | 139 x 10 ⁻⁶ | 42.7 | 0.427 |
| Bt | 1×10^{-4} | -0.470 | 118 x 10 ⁻⁶ | 51.4 | 0.514 |
| Bt | 5×10^{-4} | -0.463 | 65.1 x 10 ⁻⁶ | 73.2 | 0.732 |
| Bt | 1×10^{-3} | -0.464 | 57.9 x 10 ⁻⁶ | 76.2 | 0.762 |
| Bt | 5×10^{-3} | -0.475 | 49.2 x 10 ⁻⁶ | 79.7 | 0.797 |
| Bt | 1×10^{-2} | -0.462 | 25.8 x 10 ⁻⁶ | 89.4 | 0.894 |
| BtSO ₂ CH ₃ | 5×10^{-5} | -0.477 | 138 x 10 ⁻⁶ | 43.2 | 0.432 |
| BtSO ₂ CH ₃ | 1×10^{-4} | -0.475 | 65.5 x 10 ⁻⁶ | 73.0 | 0.730 |
| BtSO ₂ CH ₃ | 5×10^{-4} | -0.481 | 54.0 x 10 ⁻⁶ | 77.8 | 0.778 |
| BtSO ₂ CH ₃ | 1×10^{-3} | -0.476 | 32.0 x 10 ⁻⁶ | 86.8 | 0.868 |
| BtSO ₂ CH ₃ | 5×10^{-3} | -0.476 | 22.1 x 10 ⁻⁶ | 90.9 | 0.909 |
| BtSO ₂ CH ₃ | 1×10^{-2} | -0.463 | 20.5 x 10 ⁻⁶ | 91.6 | 0.916 |

Table 2. Potentiodynamic polarization result and inhibition efficiency for the corrosion of LCS in 0.1 M H_2SO_4 with and without addition of different concentration of Bt and $BtSO_2CH_3$

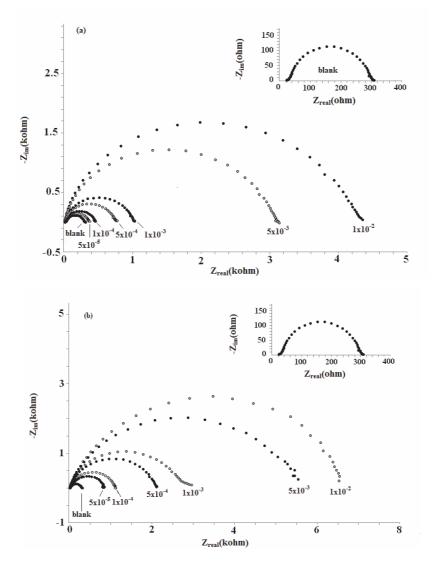


Figure 2. Impedance plots of LCS obtained in $0.1 \text{ M H}_2\text{SO}_4$ in the presence of various concentration of (a) Bt (b) BtSO₂CH₃.

Anadolu Üniversitesi Bilim ve Teknoloji Dergisi - A 12 (1) Uygulamalı Bilimler ve Mühendislik

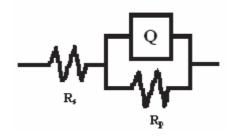


Figure 3. The electrochemical equivalent circuit according to impedance spectra.

Table 3. Impedance parameters and inhibition efficiency for the corrosion of LCS in 0.1 M H_2SO_4 with and without addition of different concentration of Bt and $BtSO_2CH_3$

| Compounds | Concentration | <i>E_{corr}</i> vs Ag/AgCl(V) | R_S (Ω) | $egin{array}{c} R_p \ (\Omega) \end{array}$ | п | $(\mu \Omega^{-1} s^n cm^{-2})$ | C_{dl} (µFcm ⁻²) | <i>IE</i> (%) | θ |
|-----------------------------------|--------------------|--|--------------------|---|------|---------------------------------|--------------------------------|---------------|-------|
| | (M) | | | | | | | | |
| Blank | - | -0.509 | 26.6 | 288.5 | 0.80 | 7.08 x 10 ⁻⁵ | 158 | - | - |
| Bt | 5×10^{-5} | -0.502 | 18.1 | 358.1 | 0.88 | 9.91 x 10 ⁻⁶ | 39.4 | 19.4 | 0.194 |
| Bt | 1×10^{-4} | -0.495 | 14.4 | 449.4 | 0.88 | 1.05 x 10 ⁻⁵ | 39.3 | 35.8 | 0.358 |
| Bt | 5×10^{-4} | -0.486 | 14.9 | 742.2 | 0.88 | 2.22 x 10 ⁻⁵ | 75.3 | 61.1 | 0.611 |
| Bt | 1×10^{-3} | -0.483 | 14.5 | 1004 | 0.88 | 2.9 x 10 ⁻⁵ | 98.5 | 71.3 | 0.713 |
| Bt | 5×10^{-3} | -0.479 | 13.8 | 3032 | 0.87 | 3.09 x 10 ⁻⁵ | 96.1 | 90.5 | 0.905 |
| Bt | 1×10^{-2} | -0.466 | 14.1 | 4193 | 0.85 | 3.48 x 10 ⁻⁵ | 94.1 | 93.1 | 0.931 |
| BtSO ₂ CH ₃ | 5×10^{-5} | -0.483 | 18.8 | 916.5 | 0.84 | 8.9 x 10 ⁻⁶ | 28.9 | 68.5 | 0.685 |
| BtSO ₂ CH ₃ | 1×10^{-4} | -0.485 | 18.5 | 1293 | 0.84 | 9.7 x 10 ⁻⁶ | 30.8 | 77.7 | 0.777 |
| BtSO ₂ CH ₃ | 5×10^{-4} | -0.496 | 18.3 | 2476 | 0.86 | 1.47 x 10 ⁻⁵ | 49.5 | 88.3 | 0.883 |
| BtSO ₂ CH ₃ | 1×10^{-3} | -0.481 | 15.3 | 3084 | 0.85 | 1.94 x 10 ⁻⁵ | 64.5 | 90.6 | 0.906 |
| BtSO ₂ CH ₃ | 5×10^{-3} | -0.489 | 13.7 | 6148 | 0.83 | 2.24 x 10 ⁻⁵ | 58.9 | 95.3 | 0.953 |
| BtSO ₂ CH ₃ | 1×10^{-2} | -0.476 | 17.9 | 6964 | 0.79 | 4.35 x 10 ⁻⁵ | 93.1 | 96.0 | 0.960 |

The inhibitor efficiency of Bt on steel surface in the presence of 1 M H_2SO_4 was obtained as 55.4% (Gomma, 1998b). Protection efficiency for steel in presence of 0.1 M H_2SO_4 containing 10^{-2} M Bt and copper cation was calculated average as 95% using potentiodynamic polarization method (Gomma, 1998c). Inhibitor efficiency of Bt for mild steel in the presence of 0.1 M HCl was found as 91.7% using the gravimetric method (Popova et al. 2007). In another work inhibitor efficiency of Bt in 0.1 M H_2SO_4 was obtained as for low and high concentration of Bt as 25.7% and 97%, respectively using potentiodynamic polarization method (Satpati and Ravindran 2008).

3.1.3 Linear Polarization Method

The polarization resistance, R_p values for LCS in sulfuric acid solution with and without of Bt and BtSO₂CH₃ were determined using linear polarization method. The obtained data from linear polarization method are presented in Table 4. The R_p and *IE*(%) values calculated from *potentiodynamic* polarization, electro-

chemical impedance spectroscopy and linear polarization methods had almost same trends.

Several electrochemical techniques are available to determine corrosion rate such as the linear polarization resistance (LPR), Tafel extrapolation and electrochemical impedance spectroscopy (EIS). The LPR and EIS techniques require B value to calculate corrosion rate from polarization resistance R_p according to the equation $i_{corr} = B/R_p$. In this equation *B* value was equal to $(\beta_a \cdot \beta_c / (\beta_a + \beta_c) 2.303)$. The Tafel extrapolation measurement, despite of its application here to evaluate corrosion rates, is time consuming and will damage the electrode surface due to the polarization over a wide potential range (Amin et al. 2010). In this study corrosion rate (CR) was determined using polarization resistance method implemented in Gamry Software Program. As can be seen from Table 4, CR values decrease with the addition of Bt and BtSO₂CH₃ and the values of CR were further decreased with the increasing Bt and BtSO₂CH₃ concentration.

| Compounds | Inhibitor Concentration (M) | E _{corr} vs Ag/AgCl (V) | <i>i_{corr}</i> (Acm ⁻²) | R_p (Ω) | <i>IE</i> (%) | Corrosion Rate (mpy) | θ |
|-----------------------------------|-----------------------------------|-------------------------------------|---|-----------------------|------------------|-------------------------|-------|
| Blank | | -0.503 | 9.269 x 10 ⁻³ | 281.1 | | 4235 | |
| Blank | 5×10^{-5} | -0.489 | 7.209×10^{-3} | 361.4 | 22.2 | 3294 | 0.222 |
| Bt | 1×10^{-4} | -0.487 | 5.828 x 10 ⁻³ | 447.0 | 37.1 | 2660 | 0.371 |
| Bt | 5×10^{-4} | -0.479 | 4.015 x 10 ⁻³ | 648.9 | 56.7 | 1835 | 0.567 |
| Bt | 1×10^{-3} | -0.477 | 2.815 x 10 ⁻³ | 925.6 | 69.6 | 1286 | 0.696 |
| Bt | 5×10^{-3} | -0.481 | 1.590 x 10 ⁻³ | 1638 | 82.8 | 726.7 | 0.828 |
| Bt | 1×10^{-2} | -0.467 | 647.1 x 10 ⁻⁶ | 4026 | 93.0 | 295.7 | 0.930 |
| BtSO ₂ CH ₃ | 5×10^{-5} | -0.487 | 4.353 x 10 ⁻³ | 598.6 | 53.0 | 1980 | 0.530 |
| BtSO ₂ CH ₃ | 1×10^{-4} | -0.485 | 2.540 x 10 ⁻³ | 1026 | 72.6 | 1160 | 0.726 |
| BtSO ₂ CH ₃ | 5×10^{-4} | -0.480 | 1.926 x 10 ⁻³ | 1353 | 79.2 | 880.0 | 0.792 |
| BtSO ₂ CH ₃ | 1×10^{-3} | -0.480 | 1.021 x 10 ⁻³ | 2551 | 88.9 | 466.7 | 0.889 |
| BtSO ₂ CH ₃ | 5×10^{-3} | -0.483 | 694.3 x 10 ⁻⁶ | 3752 | 92.5 | 317.3 | 0.925 |
| BtSO ₂ CH ₃ | 1×10^{-2} | -0.473 | 412.5 x 10 ⁻⁶ | 6315 | 95.5 | 188.5 | 0.955 |

Table 4. Linear polarization result for the corrosion of LCS in 0.1 M H₂SO₄ with and without addition of different concentration of Bt and BtSO₂CH₃

3.1.4 Adsorption

The surface coverage ratios (θ) for various concentrations of the inhibitors in H₂SO₄ were evaluated using potentiodynamic polarization, electrochemical impedance measurements and linear polarization resistance methods. The obtained experimental data for the studied inhibitors have been applied to different adsorption isotherm equation. The best correlations have been obtained from each method and the results obey Temkin and Langmuir adsorption isotherm equations for Bt and BtSO₂CH₃, respectively (Hosseini et al. 2003; Fouda et al. 2010).

The adsorption isotherms were plotted for Bt and BtSO₂CH₃. The equilibrium constants of adsorption (K_{ads}) were calculated from obtained straight lines in these plots. The equilibrium constants of adsorption are related to the free energy of adsorption (ΔG_{ads}) by;

$$K_{abs} = \frac{1}{55.5} e^{-\Delta G_{abs}/RT}$$

where 55.5 is the molar concentration of water in the solution, *T* is the temperature, and *R* is the universal gas constant. The calculated values of K_{ads} and ΔG_{ads} are given Table 5.

$$\theta = \left[1 - \frac{R_p(blank)}{R_p(inh)}\right]$$

The low and negative values of ΔG_{ads} indicate the spontaneous adsorption of inhibitors on the surface of steel. The negative values of ΔG_{ads} also suggest the strong interaction of the inhibitor molecules onto the steel surface (Quraishi et al. 2001; Elachouri et al. 1996).

4. CONCLUSIONS

- Bt and BtSO₂CH₃ show inhibiting properties for the corrosion of LCS in 0.1 M H₂SO₄ solutions.
- The inhibition efficiency increases with increasing Bt and BtSO₂CH₃ concentrations.
- The inhibition efficiency Bt and BtSO₂CH₃ using *potentiodynamic* polarization, linear polarization resistance and electrochemical impedance spectroscopy methods showed similar trends.
- The adsorption of Bt and BtSO₂CH₃ on the LCS surface obeys Temkin and Langmuir adsorption isotherms.

| Inhibitör | Po | tantiodyn | otantiodynamic polarisatior | ntion | T | ineer polar. | Jineer polarisation resistance | tance | Electroc | shemical im | Electrochemical impedance spectroscopy | troscopy |
|-----------------|--------|-----------|-----------------------------|----------------|----------|--------------|--------------------------------|----------------|----------|-------------|--|----------------|
| | | | | | | ن ا | | | | 9 | | |
| | R^2 | f | K_{ads} | $\Delta G(kJ)$ | R^2 | f | K_{ads} | $\Delta G(kJ)$ | R^2 | f | K_{ads} | $\Delta G(kJ)$ |
| Bt | 0.9238 | 12500 | 16043.75 | -33.940 | 0.9855 | 10000 | 15333 | -33.826 | 0.9761 | 10000 | 13602 | -33.531 |
| BtSO,CH, 0.9634 | 0 9634 | 0 4961 | 10039.08 | -32,778 | 0 8621 | 0 2,978 | 0 2978 9693 69 -36 692 | -36.692 | 0 9452 | 0 2399 | 9937 14 | -32,754 |

Table 5. Thermodynamic parameters of adsorption obtained each methods for Bt and BtSO₂CH₃ on LCS in 0.1 M H₂SO₄ solutions.

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