



## KINETIC AND THERMODYNAMIC STUDY OF THE CORROSION RESISTANCE IMPROVEMENT OF CARBON STEEL BY GREEN CORROSION INHIBITOR IN SIMULATED SEA WATER.

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### Abstract

The use of green corrosion inhibitor extracted from *Thaumatococcus danielli* Plant (TDE) was investigated on carbon steel in simulated sea water using gravimetric (weight loss) and Scanning electron microscopy (SEM) methods at different temperatures. Increased activation energy values of the corrosion process results from a decrease in inhibition efficiency as temperature rises, which points to a physical adsorption mechanism. The value of activated enthalpy ( $\Delta H^*$ ) and entropy ( $\Delta S^*$ ) suggest endothermic and spontaneous adsorption of the *Thaumatococcus danielli* extract (TDE) constituent on the steel surface. The kinetic study of the corrosion inhibition of carbon steel in simulated sea water in the absence and presence of *Thaumatococcus danielli* extract (TDE) obeyed first order reaction kinetic. An examination of the surface morphology after the addition of the inhibitor TDE reveals some improvements on the steel surface, this suggest that *Thaumatococcus danielli* extract (TDE) is primarily responsible for the corrosion inhibition of the carbon steel.

**Keywords:** Simulated Sea Water, Carbon Steel, Kinetic, Thermodynamic, SEM

### 1. Introduction

Corrosion is one of the most significant limitations to the use of steel and other materials in industrial applications (Emori *et al.*, 2018). According to estimates, the cost of corrosion is in the range of 3.5% of gross national product of developed countries (Ikpi *et al.*, 2017a). With numerous methods being suggested, corrosion control has

proven to be an expensive procedure. Researchers have identified a number of workable methods to prevent corrosion, including the modification of materials, solutions modification, operations improvements and the application of corrosion inhibitors and coating (Deyab and Guibal, 2020; Hanza *et al.*, 2016; Asri *et al.*, 2017; Ekerenam *et al.*, 2017; Escrivà-Cerdan

*et al.*, 2019; Barmatov *et al.*, 2016; Deyab, 2015). Due to its low overall cost and high returns on investment, the application of corrosion inhibitors is acknowledged as the most effective and efficient method. Despite the development of a variety of corrosion inhibitors, compounds that are environmentally friendly and biodegradable have been the preferred options. As a result, there have been increasingly carefully studies on the use of natural products as efficient inhibitors in a variety of environments and applications (Abeng *et al.* 2013; Abeng and Idim 2018; Abeng *et al.* 2021; Okafor *et al.* 2010; Mobin and Rizvi, 2017; Wang *et al.*, 2019; Deyab and Guibal, 2020; Anadebe *et al.* 2021). The majority of preferred organic compounds have aromatic rings, heteroatoms like N, S, O, and P, and typically contain pi-electrons in their structures and these are mostly chosen. This is due to the ease with which they interact with the surfaces of metals through adsorption using these functional groups, ultimately providing the metals with protection against aggressive environments. The type of corrosion medium, the chemical structure of the inhibitor compound, and the surface characteristics of the metal all have an impact on how an inhibitor compound interacts with the surface of a metal (Emori *et al.*, 2020). The leaves of *Thaumatococcus daniellii* extract (TDE) has been tested as a locally source corrosion inhibitor for preventing corrosion of carbon steel used in simulated sea water by chemical method (weight loss). The common name of *Thaumatococcus daniellii* leaves is called moimoi leaves, some called it food wrapper. *Thaumatococcus daniellii* is one of the plants that has long been used in traditional

medicine. To the best of our knowledge, the plant *Thaumatococcus daniellii* leaves extract has not been the subject of any corrosion research. According to Ushie *et al.*, 2022; Hamid *et al.*, 2017; Ayodeji *et al.*, 2016, the leaves of *Thaumatococcus daniellii* contains alkaloids, saponins flavonoid, tannins and phenols these constituents are the properties that are found in good corrosion inhibitors because there contain some heteroatoms like carbon, oxygen and nitrogen in them. Therefore the objectives of this research paper is to investigate the kinetics and thermodynamic study of corrosion resistance improvement of carbon steel used in simulated sea water by *Thaumatococcus daniellii* as a green corrosion inhibitor.

## 2. Materials and methods

### 2.1. Metal specimen preparation

Gravimetric experiment was performed on a carbon steel (A570 Gr40) specimen of the composition; 0.25 %C, 1.60 % Mn, 0.04 % P, 0.05 % S, 0.05 % Si, and 98.01 % Fe. The surface of the specimens was prepared by using sandpaper with grits of 600 and 320, washed in deionized water, dried and used immediately after the pre-treatment (Ikpi and Abeng, 2020).

### 2.2 Corrosion inhibitor Preparation

*Thaumatococcus daniellii* leaves were purchased on the spot in Marian Market in Cross River State, Nigeria. After being dried in the sun for two days to remove moisture, the leaves were ground into a fine powder and then filtered to produce extremely smooth leaf particles. The fine powder was thoroughly soaked in methanol solution for 24 hours with periodic stirring to create a homogenous solution. The extract was then

recovered through filtration in the form of paste. The filtrate was then subjected to an evaporation process in order to remove the extra alcohol (Sedik, *et al.*, 2020). The filtrate was used as an inhibitor in its purest form. The extract was diluted in one liter of prepared simulated sea water and allowed to stand for 24 hours before been used. The

stock solution inhibitor test solution was prepared for different concentration of 0.1, 0.5, and 2.0 g/L, 3.0 g/L using dilution formula Fouda *et al.*,2020; Akinbulumo *et al.*, 2020). The simulated sea water use in this study was prepared base on the ASTM D1141-98 International standard formula presented in Table 1. (Axelrod, 2005).

Table 1. Formula for artificial seawater ASTM D1141-98 composition.

| SALT   | CONCENTRATION (g/L) |
|--|---------------------|
| Sodium Chloride (NaCl)                             | 24.53               |
| Sodium Sulphate (Na <sub>2</sub> SO <sub>4</sub> ) | 4.09                |
| Potassium Chloride (KCl)                           | 0.595               |
| Sodium bicarbonate (NaHCO <sub>3</sub> )           | 0.201               |
| Boric acid (H <sub>3</sub> BO <sub>3</sub> )       | 0.027               |
| Magnesium Chloride (MgCl <sub>2</sub> )            | 5.20                |
| Calcium Chloride (CaCl <sub>2</sub> )              | 1.16                |
| Strontium Chloride (SrCl <sub>2</sub> )            | 0.0025              |
| Sodium Fluoride (NaF)                              | 0.003               |
| Potassium Bromide KBr                              | 0.101               |
| Water (H <sub>2</sub> O)                           | 988.968             |

### 2.3 Gravimetric measurement

Measurements of weight loss were used to determine the impact of immersion time and temperature. The experiments on the effects of temperature were conducted at 313, 323 and 333 K. Gravimetric tests were performed using test coupons with the following dimensions: 1 cm x 1 cm x 1 cm. Glass hooks and rods were used to suspend the pre-cleaned and weighed coupons in the beaker containing the test solutions. Tests were carried out in 300 mL of the test solutions under total immersion conditions. The coupons were taken out at 24-hour

intervals, repeated 120 hours, scrubbed with bristle brushes, washed, dried, and weighed to determine weight loss over time. The difference between the weight of the coupons at a given time and their initial weight was assumed to be the weight loss. To ensure accurate, reproducible results, every test was performed twice. Each experiment's average values were gathered and used in the calculations that followed. Inhibitors of various concentrations were added to 300 mL of simulated sea water, where the carbon steel specimen was submerged in duplicate. Equations below

were used to calculate the percentage inhibition efficiency (IE) and corrosion rate (CR) of carbon steel (Aralu *et al.*, 2021; Emori *et al.*, 2020; Di *et al.*, 2022).

$$IE \% = \left(1 - \frac{W_{blank}}{W_{inh}}\right) \quad (1)$$

$$CR (gh^{-1}cm^{-2}) = \frac{\Delta W}{AT} \quad (2)$$

where  $W_{inh}$  is the weight loss of carbon steel in the presence of inhibitor and  $W_{blank}$  is the

weight loss of carbon steel in absence of inhibitor.

### 3. Results and discussion

#### 3.1 Effect of inhibitor concentration and time

As a function of concentration and time in the presence of *Thaumatococcus danielli* extract (TDE), the variation of weight loss, obtained from a weight loss experiment of carbon steel in simulated sea water at various temperatures is depicted in Figure 1.

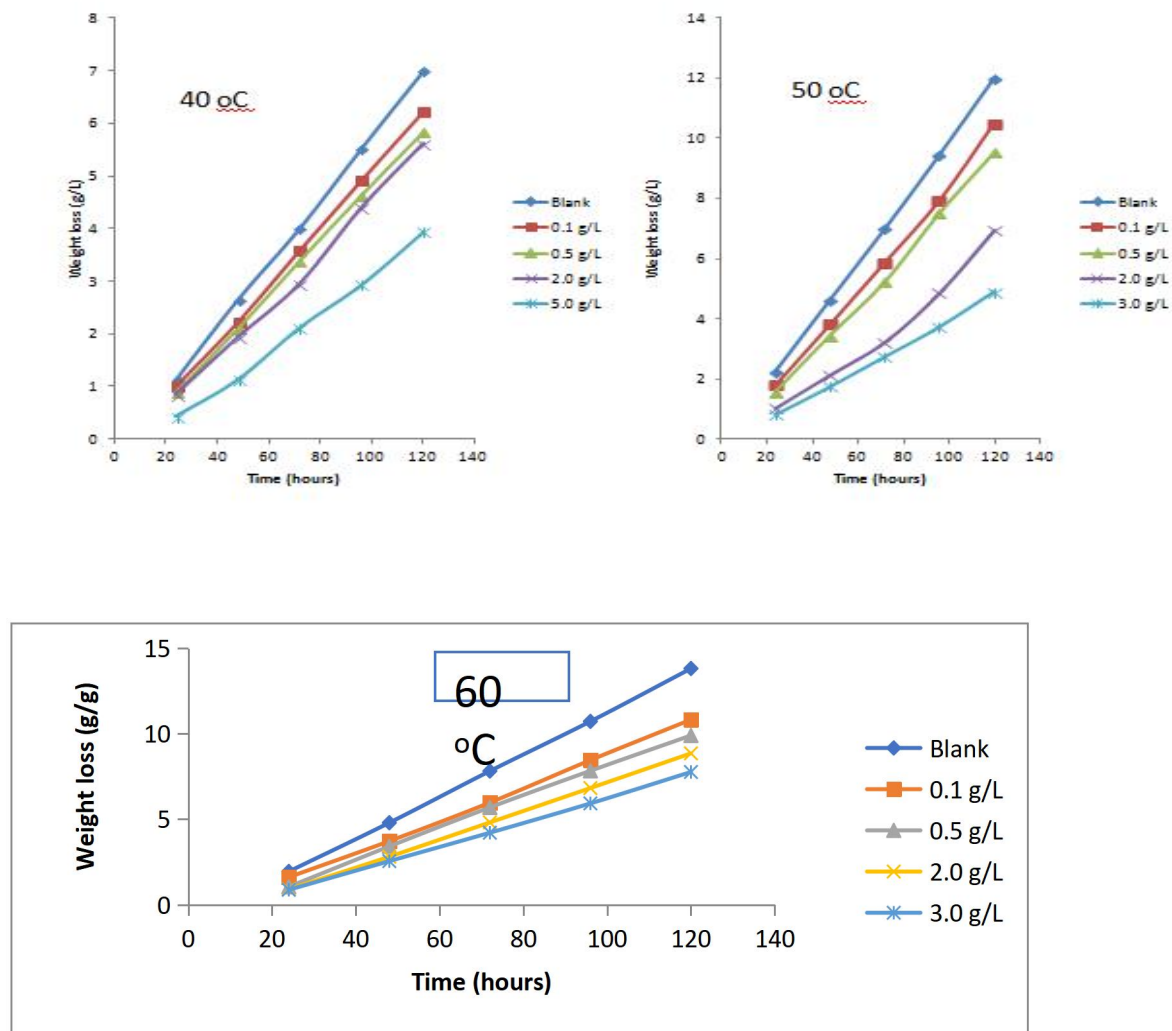


Figure 1. Variation of weight loss with time for the corrosion of carbon steel in simulated sea water at different temperatures.

It is clearly seen in the Figure 1 above that weight loss of the carbon steel specimen immersed in simulated sea water without TDE is high compare to the carbon steel specimen immersed in simulated sea water with TDE, this revealed that study plant extract improved the corrosion resistance of the studied carbon steel in simulated sea water.

Table 2 displays that the inhibition efficiency of carbon steel decreased over time but increased with increasing TDE inhibitor concentrations, while the corrosion rate of the study steel increases with time and decreases with addition of the inhibitor concentration. This behavior might be explained by an increase in inhibitor adsorption on the steel surface or at the solution-solution interface as the concentration increases (Ijuo *et al.*, 2016). As exposure time increased, inhibition efficiency decreased because the aggressive action of the aqueous medium became increasingly more noticeable than the adsorbed inhibitor (Dehghan *et al.*, 2019a; Asad *et al.*, 2019; Dehghani *et al.*, 2019b). This result agrees with the preliminary reports (Ijuo *et al.*, 2016).

### 3.1.1 Kinetic Study

The rate constant (K) of the carbon steel was obtained from the slope of the plots of  $\ln \frac{W_f}{W_i}$  against time in hours (Figure 2) and the half-life of study metal was calculated from equation 3.

$$t_{1/2} = \frac{0.693}{k_1} \quad (3)$$

The Values of rate constant parameters (K) listed in Table 3, decreases as the concentration of the inhibitor increased and the half-life of the metal increases as the concentration of the inhibitor increased this is due to the adsorption of the TDE on the steel surface. The plots of corrosion of carbon steel in simulated sea water in the absence and presence of TDE revealed a linear variation which supports a first order reaction kinetic. This result is in line with the report of Ikpi *et al.*, 2017b; Elmsellem *et al.*, 2019.

### 3.2 Effect of concentration and temperature

The effect of temperature on the corrosion of carbon steel is illustrated in Table 2 above, according to the findings, inhibition efficiency increased as inhibitor concentration increased but decreased as temperature increased. In the other way round the corrosion rate decreases with increased in inhibitor concentration and increased as the temperature increases. This is additional proof that TDE effectively inhibits carbon steel corrosion in simulated sea water at lower temperature.

The increase in corrosion rate and decrease in inhibition efficiency with increasing temperature has also been attributed to the nature of adsorption, in which the inhibitor is physically adsorbed at lower temperatures while chemisorption is preferred at higher temperatures (Ijuo *et al.*, 2016; Ihebrodike *et al.*, 2010; Bangeraet *et al.*, 2022; Karkiet *et al.*, 2021). Physiosorption was preferred in our study. Table 2 summarizes the inhibition efficiency and corrosion rate at various temperatures.

Table 2. Inhibition efficiency and corrosion rates ( $\times 10^{-3} \text{ gh}^{-1}\text{cm}^{-2}$ ) of carbon steel in various TDE concentrations at different time interval.

| Tem<br>oC               | System  | 24       | 48       | 72       | 96       | 120      |
|-------------------------|---------|----------|----------|----------|----------|----------|
| % Inhibition efficiency |         |          |          |          |          |          |
| 40 °C                   | 0.1 g/L | 19.86    | 17.5     | 15.97    | 16.06    | 12.53    |
|                         | 0.5 g/L | 29.97    | 25.61    | 25.19    | 20.47    | 20.67    |
|                         | 2.0 g/L | 54.61    | 54.05    | 53.91    | 48.57    | 42.18    |
|                         | 3.0 g/L | 63.74    | 62.35    | 60.89    | 60.63    | 59.25    |
| 50 °C                   | 0.1 g/L | 11.36    | 16.37    | 10.46    | 10.95    | 10.95    |
|                         | 0.5 g/L | 21.04    | 20.68    | 15.32    | 16.02    | 16.53    |
|                         | 2.0 g/L | 25.31    | 27.23    | 26.31    | 20.38    | 19.62    |
|                         | 3.0 g/L | 62.52    | 57.82    | 47.29    | 47.07    | 43.73    |
| 60 °C                   | 0.1 g/L | 17.11    | 22.39    | 23.58    | 21.14    | 21.64    |
|                         | 0.5 g/L | 45.99    | 28.79    | 27.2     | 26.94    | 28.33    |
|                         | 2.0 g/L | 53.28    | 41.13    | 38.42    | 36.34    | 35.91    |
|                         | 3.0 g/L | 54.93    | 46.76    | 46.11    | 44.71    | 43.79    |
| Corrosion Rate          |         |          |          |          |          |          |
| 40 °C                   | Blank   | 7.819444 | 9.180556 | 9.24537  | 9.574653 | 9.695833 |
|                         | 0.1 g/L | 6.930556 | 7.677083 | 8.277778 | 8.526042 | 8.633333 |
|                         | 0.5 g/L | 6.173611 | 7.28125  | 7.828704 | 8.039931 | 8.093056 |
|                         | 2.0 g/L | 5.840278 | 6.680556 | 6.8125   | 7.623264 | 7.793056 |
|                         | 3.0 g/L | 2.930556 | 3.871528 | 4.872685 | 5.067708 | 5.455556 |
| 50 °C                   | Blank   | 15.59028 | 16.06597 | 16.15972 | 16.39757 | 16.66389 |
|                         | 0.1 g/L | 12.49306 | 13.25347 | 13.5787  | 13.76389 | 14.575   |
|                         | 0.5 g/L | 10.91667 | 11.95139 | 12.08796 | 13.03993 | 13.21944 |
|                         | 2.0 g/L | 7.076389 | 7.381944 | 7.446759 | 8.432292 | 9.634722 |
|                         | 3.0 g/L | 5.652778 | 6.048611 | 6.319444 | 6.454861 | 6.788889 |
| 60 °C                   | Blank   | 13.51389 | 16.65278 | 18.09722 | 18.61285 | 19.18333 |
|                         | 0.1 g/L | 11.20139 | 12.92361 | 13.8287  | 14.67708 | 15.03194 |
|                         | 0.5 g/L | 7.298611 | 11.85764 | 13.17407 | 13.59722 | 13.74722 |
|                         | 2.0 g/L | 6.3125   | 9.760417 | 11.14352 | 11.84722 | 12.29444 |
|                         | 3.0 g/L | 6.090278 | 8.864583 | 9.752315 | 10.28993 | 10.78194 |

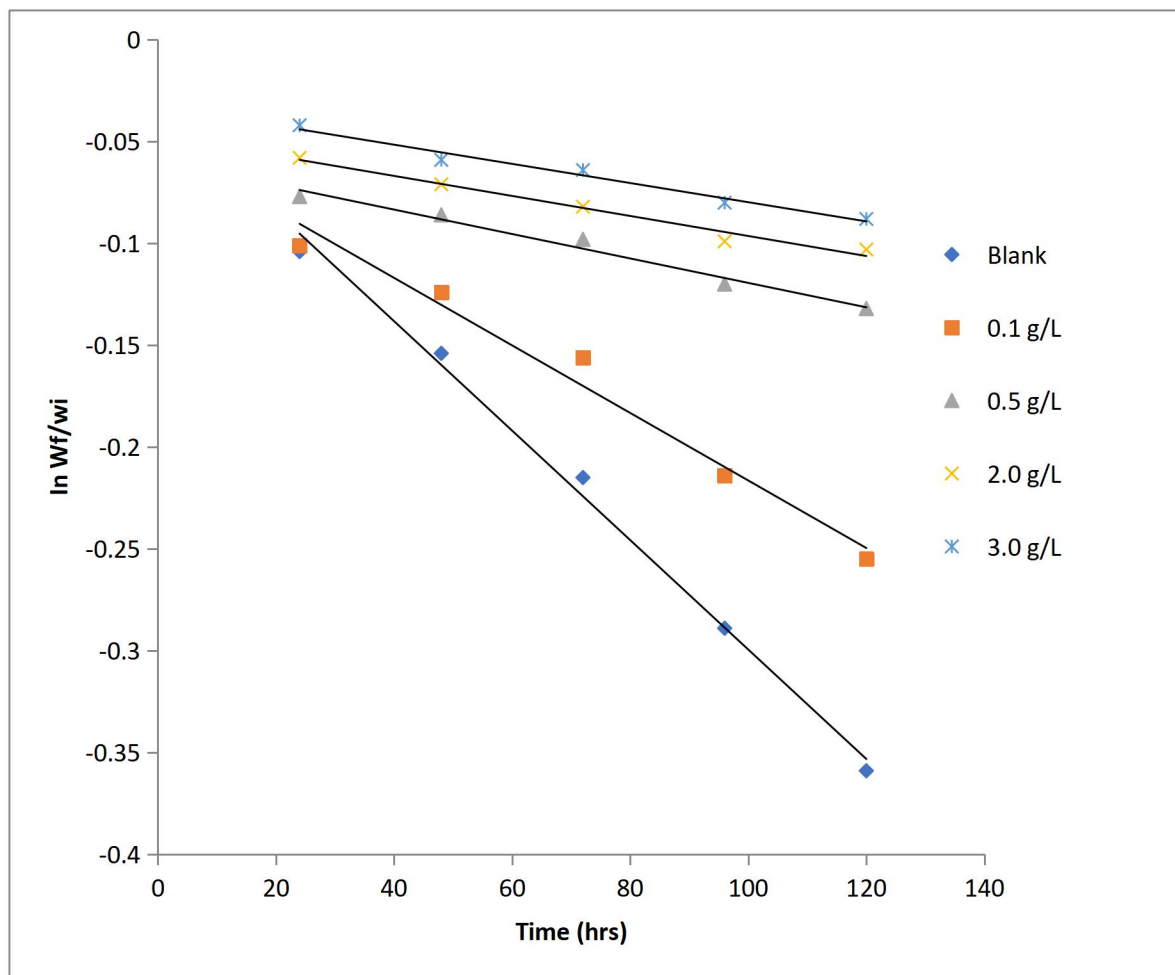


Figure 2. Plots of  $\ln \frac{W_f}{W_i}$  against time in hours for corrosion of carbon steel in simulated sea water with and without TDE.

Table 3. Kinetic Parameters for the corrosion inhibition of carbon steel in simulated sea water with and without TDE.

| Conc. (g/L) | K       | $t_{1/2}$ (hrs) | $R^2$ | Qads (kJmol <sup>-1</sup> ) |
|-------------|---------|-----------------|-------|-----------------------------|
| Blank       | 0.002   | 347             | 0.994 |                             |
| 0.1         | 0.001   | 693             | 0.975 | 5.60                        |
| 0.5         | 0.00005 | 13860           | 0.978 | 2.99                        |
| 2.0         | 0.00004 | 17325           | 0.977 | 3.86                        |
| 3.0         | 0.00003 | 23100           | 0.976 | 2.05                        |

### 3.2.1 Thermodynamic consideration

Following the effect of temperature on the corrosion of carbon steel in simulated sea

water in the absence and presence of TDE in various concentrations, it was revealed that the corrosion rate increased exponentially with rise in temperature. According to Abeng *et al.* (2013), the activation energies ( $E_a$ ) were calculated using the Arrhenius equation (equation 4).

$$CR = A \exp\left(\frac{-E_a}{RT}\right) \quad (4)$$

Further derivation of equation 4 gives equation 5 where R is the universal gas constant, CR is the corrosion rate and A is the pre-exponential factor.

$$\log CR = \log A - \frac{E_a}{RT} \quad (5)$$

Figure 3a, illustrates the relationship between the logarithm of corrosion rate and the reciprocal of absolute temperature. Table 4 illustrate the calculated values of apparent activation energy ( $E_a^*$ ) of corrosion inhibition of carbon steel in simulated sea water in absence and presence of TDE, these

values were obtained from the slope of  $\log CR$  versus  $1/T$  in Figure 3a and the expression of the slope is  $-E_a/R$  according to equation 5. From our results it is clearly seen that addition of the extract concentration in grams per liter increases the values of activation energy ( $E_a^*$ ) for the corrosion of carbon steel in simulated sea water, indicating that the study plant extract TDE is ineffective at higher temperatures but very effective in lower temperature, which would correspond to the observed decrease in inhibition efficiency with temperature. The trend in activation energy ( $E_a^*$ ) proposed physical adsorption mechanism for the adsorption of the TDE. (Abeng *et al.*, 2017; Bangera and Alva, 2020). The Eyring transition state equation provided some additional thermodynamic activation parameters, including the enthalpy change of activation ( $\Delta H^*$ ) and entropy change of activation ( $\Delta S^*$ ).

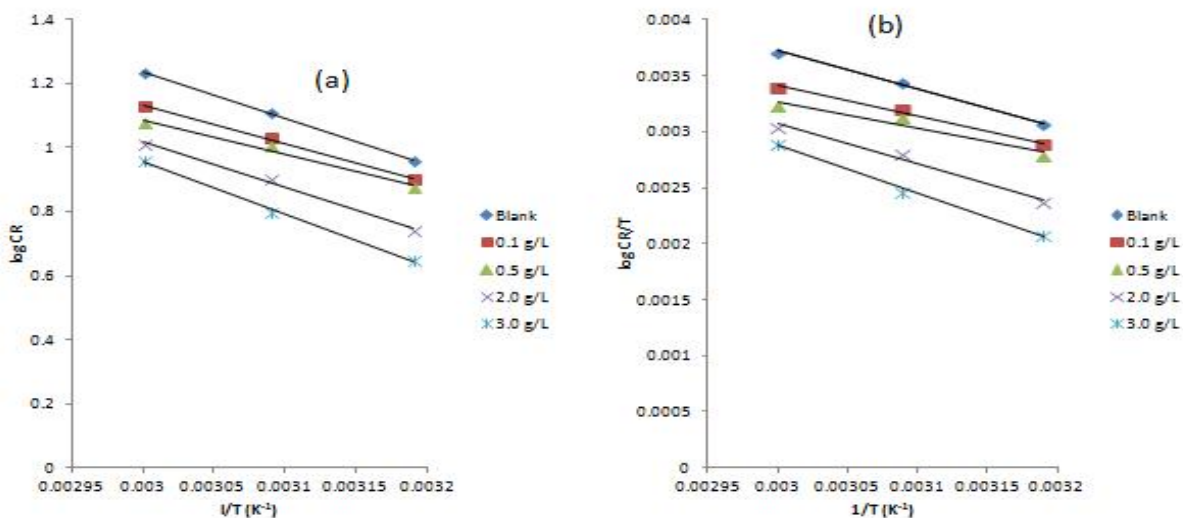


Figure 3. (a) Arrhenius and (b) Eyring transition state plots for the corrosion inhibition of carbon steel in simulated sea water in absence and different concentration of TDE.



Eyring transition state equation is given as equation 6. In this equation,  $h$  stands for the Planck constant,  $N$  for the Avogadro number,  $\Delta S^*$  for activation entropy, and  $\Delta H^*$  for activation enthalpy. These thermodynamic parameters were obtained from the plots of  $\log CR/T$  versus  $1/T$  shown in Figure 3b.

$$CR = \frac{RT}{Nh} \exp\left(\frac{\Delta S^*}{R}\right) \exp\left(\frac{-\Delta H^*}{RT}\right) \quad (6)$$

The values of  $\Delta H^*$  and  $\Delta S^*$  were calculated from a straight line with a slope of  $-\Delta H^*/R$  and intercept of  $\ln(R/Nh + \Delta S^*/R)$ . The results shown in Table 4 demonstrated that the process of dissolving the metal in the simulated sea water in the absence and presence of TDE reflected endothermic, as indicated by the positive enthalpy values (Go *et al.*, 2020).

Table 4. Parameters of thermodynamics for the adsorption of TDE on the carbon steel surface.

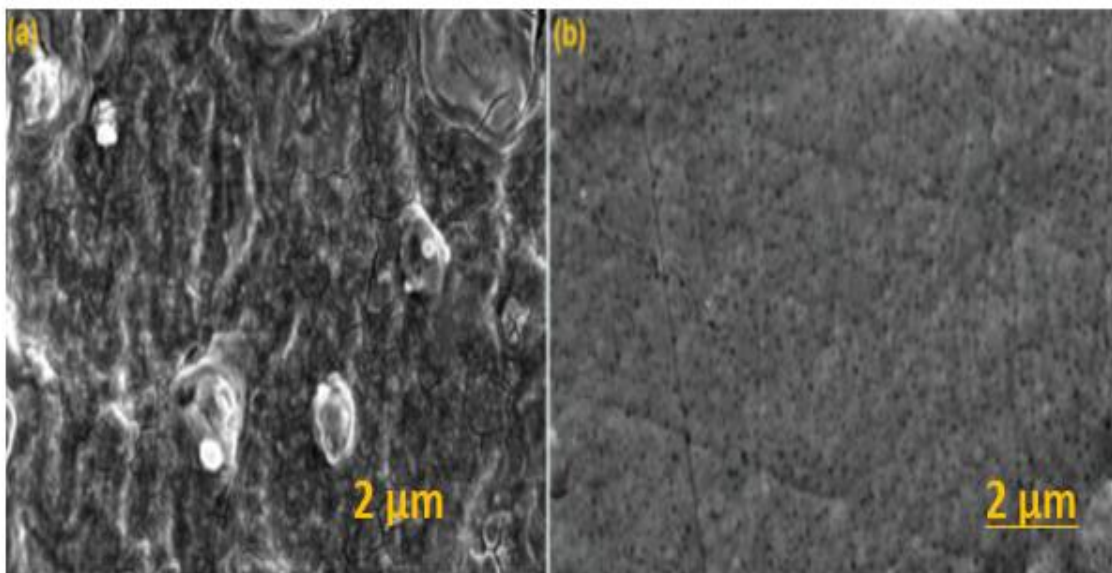
| System  | Ea* (KJmol <sup>-1</sup> ) | R <sup>2</sup> | ΔH*(J mol <sup>-1</sup> ) | R <sup>2</sup> | ΔS* (kJmol <sup>-1</sup> ) |
|---------|----------------------------|----------------|---------------------------|----------------|----------------------------|
| Blank   | 27.9                       | 1.000          | 65.3                      | 0.998          | 13.9                       |
| 0.1 g/L | 23.0                       | 0.998          | 51.6                      | 0.993          | 11.5                       |
| 0.5 g/L | 20.5                       | 0.977          | 44.8                      | 0.942          | 10.3                       |
| 2.0 g/L | 27.2                       | 0.994          | 67.5                      | 0.984          | 13.6                       |
| 3.0 g/L | 31.6                       | 0.996          | 82.5                      | 0.998          | 15.8                       |

Additionally, the activation energy entropies for the study plant extract were positive, demonstrating that the activation complex represents association steps and that the reaction was feasible and spontaneous.

### 3.3 Carbon steel surface characterization

Scanning electron microscopy analysis is a useful technique for examining the surface morphology of steel surfaces after it has been exposed to various media. This method is especially helpful for describing how a surface protective organic layer develops to allow for high levels of inhibition resistance (Diet *et al.*, 2022; Aralu *et al.*, 2021;

Abeng *et al.*, 2021; Abenget *et al.*, 2020a). The surface morphology of carbon steel surfaces that have been processed and immersed for 24 hours in two different solutions, one without any TDE inhibitor and one with 3.0 g/L of TDE inhibitor is shown in Figure 4, and Figure 4 demonstrates that the two SEM micrographs significantly differ from one another. Without inhibitors, as shown in Figure 4a, the carbon steel surface is severely corroded and contains internal corrosion damage as a result of the rapid corrosion attack in the simulated seawater; as a result, the carbon steel surface becomes irregular and rough.



Fig

ure 4 (a) and (b). Scanning electron microscopy SEM of carbon steel specimens after 24 hours immersion in simulated sea water in (a) absence and (b) presence of 3.0 g/L of TDE at 60°C.

Figure 4b, on the other hand, shows that the presence of the TDE inhibitor significantly reduces the damage on the carbon steel surface, implying that the carbon steel surface morphology is more protected due to TDE adsorption onto the carbon steel surface (Bangera *et al.*, 2022; Abeng *et al.*, 2020b). This result confirms that the studied *Thaumatococcus danielli* extract TDE has significantly improved the resistance of the carbon steel used in simulated sea water.

#### 4. Conclusion

This study yielded the following key findings. Increased activation energy of the corrosion process results from a decrease in inhibition efficiency as temperature rises, which points to a physical adsorption mechanism. An examination of the surface morphology after the addition of the inhibitor reveals some improvements on the steel surface. The *Thaumatococcus danielli* extract is primarily responsible for the

corrosion inhibition of the carbon steel. The kinetic study of the corrosion inhibition of carbon steel in simulated sea water in the absence and presence of *Thaumatococcus danielli* extract TDE obeyed first order reaction kinetic. The values of activated enthalpy ( $\Delta H^*$ ) and entropy ( $\Delta S^*$ ) suggest endothermic and spontaneous adsorption of the *Thaumatococcus danielli* extract TDE constituent on the steel surface.

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