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Full Length Article

Physicochemical studies on the inhibitive properties of a 1,2,4-triazole Schiff's base, HMATD, on the corrosion of mild steel in hydrochloric acid

P. Rugmini Ammal, M. Prajila, Abraham Joseph*

Department of Chemistry, University of Calicut, Calicut University, P O-673 635, Kerala, India

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ABSTRACT

The efficiency of 4-(4-hydroxy-3-methoxy benzylidene amino)-4-H-1,2,4-triazole-3, 5-dimethanol, HMATD, as corrosion inhibitor for mild steel in 0.5 M HCl has been determined by weight loss measurements and electro analytical methods. The influence of various parameters such as temperature, inhibitor concentration on the efficiency has been studied. The electrochemical impedance spectroscopic measurements revealed the inhibition action of HMATD by reducing the charge transfer through metal solution interface. Polarization curves indicate the mixed type behaviour of HMATD. The inhibitor molecule functions by blocking the active sites on metal surface by adsorption and which obeys the Langmuir adsorption isotherm. Various kinetic and thermodynamic parameters have also been calculated.

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1. Introduction

The extensive use of acid solutions in the process of metal cleaning and the resulting metal corrosion has posed challenging tasks for researchers in materials engineering in view of satisfying environmental requirements and for industrial facilities [1]. The driving force for the corrosion of metals is their increasing tendency to remain in the native form of oxides compared to the highly energetic pure metallic form. Corrosion of metals in mineral acids is a terrible waste both of money and resources. Out of the various methods practised for combating aqueous and process industry, the use of corrosion inhibitors has attained an interesting concern among the researchers. The structural functionalities of inhibitors and their specific interaction with the corrosion active centres on the metal surface urged the research community into the development of new inhibitors. It has been found that organic compounds with structural features like hetero atoms, with lone pair of electrons like N, S, O, P etc [2–4], the presence of multiple bonds and aromatic rings [5–6] can effectively act against the attack of corrosive media by adsorption through these structural features forming a protective film on the metal surface [7–8]. These

inhibitors act either by decelerating the anodic metal dissolution reaction or the cathodic hydrogen evolution, or both [9].

Schiff bases are compounds with RC = NR' (azomethine) group produced by the condensation of an amine with a carbonyl compound. These classes of compounds were widely used in many fields of chemical production and scientific research as chelating agents [10–12], biologically active agents [13–15], stabilizers, catalytic reagents [16,17] etc. Among various classes of N-containing heterocyclics, Schiff's bases were tested to offer very good corrosion inhibition efficiency for mild steel [18–21], aluminium [22,23], copper [24,25] etc since they possess the structural features required for corrosion inhibition. The process of synthesis of Schiff's base is relatively simple and one can introduce groups with hetero atoms and can tune the desired properties of molecules. Some of the researchers reported the enhanced corrosion inhibition of Schiff's bases compared to their parent amine and/or aldehyde due to the introduction of azomethine group (-C = N-) in the molecule [26]. Ramya et al. reported the corrosion inhibition of a novel 1,2,4-triazole and vanilline based Schiff base, HMATD, for copper corrosion in nitric acid [27]. The HMATD molecule contains various anchoring sites suitable for surface bonding like azomethine nitrogen, aromatic rings, oxygen atom etc.

The present work intends to investigate the function of 1,2,4-triazole based Schiff base 4-(4-hydroxy-3-methoxy benzylidene amino)-4-H-1,2,4-triazole-3, 5-dimethanol, HMATD, as an inhibi-

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* Corresponding author.

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tor to mitigate mild steel corrosion in hydrochloric acid at various temperatures by weight loss and electrochemical methods.

2. Experimental

2.1. Inhibitor molecule

The inhibitor, HMATD was 4-(4-hydroxy-3-methoxy benzylidene amino)-4-H-1,2,4-triazole-3, 5-dimethanol, HMATD, whose structure is given in Fig. 1. This was synthesized by the condensation of 1,2,4-triazole precursor 4-amino-4-H-1,2,4-triazole-3,5-dimethanol (ATD) with vanilline using alcohol as the solvent [27] described in Scheme 1 and was characterized by various physicochemical methods like IR and NMR [27] and the respective spectra are given in Figs. 2 and 3. The prominent FTIR and ^1H NMR assignments of the molecule are as follows: IR: 3413 cm^{-1} ν (O–H), 1590 cm^{-1} ν (C=N), 1377 cm^{-1} δ (phenolic O–H), 1284 cm^{-1} ν (C–O), 1027 cm^{-1} ν (N–N). ^1H NMR DMSO- d_6 : δ 10 (phenolic OH), δ 8.8 (CH=N), δ 7.5–6.9 (aromatic protons), δ 5.6 (OH), δ 4.6 (CH_2), δ 3.8 (OCH_3).

2.2. Materials and method

The material used for corrosion studies was mild steel having the composition (wt%): C (0.20%), Mn (1%), P (0.03%), S (0.02%) and Fe (98.75%), which was mechanically press cut into specimens of dimension $2 \times 1.8\text{ cm}^2$ in area and was used for the weight loss measurements. The electrochemical studies were conducted using the above said metal coupons as working electrode with an exposed area of 1 cm^2 . The specimens were polished using various grades of emery paper, washed with distilled water, degreased in acetone, ethanol and dried at room temperature as per ASTM standard G-1-72.

The corrosive medium selected was 0.5 M HCl prepared from reagent grade HCl (Merck) by dilution with double distilled water. All the studies were conducted in aerated DMSO medium under standard conditions.

2.3. Weight loss method

Gravimetric measurements were performed on mild steel coupons cleaned as per ASTM standard immersed in 0.5 M HCl solution with and without inhibitors for specific immersion time. The coupons were weighed before and after immersion in an analytical balance. From the weight loss corrosion rate (CR) and inhibition efficiency (IE) were calculated.

2.4. Electrochemical studies

Electrochemical studies were carried out using a computer controlled electrochemical work station ACM Gill AC Sequencer (model no: 1475). A three electrode system consisting of mild steel as working electrode, a platinum electrode as counter electrode

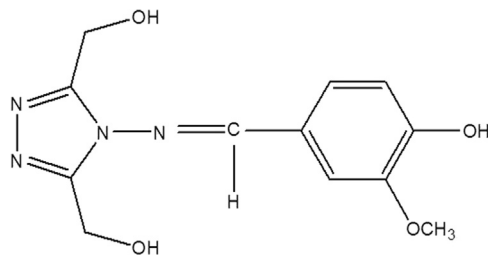


Fig. 1. Structure of inhibitor molecule.

and a saturated calomel electrode as reference electrode was used. The potentiodynamic polarization curves were recorded by polarizing the electrode first cathodically by -250 mV and then $+250\text{ mV}$ anodically with respect to the open circuit potential at a scan rate of 1 mV/s . Electrode potentials were measured with respect to SCE. As reported in the literature it was believed that the strength of the acidic media will not be drastically affected by the corrosion reactions during the study, especially in the presence of the inhibitors [28]. Prior to polarization studies impedance studies were performed on the same working electrode in a frequency range of 10 kHz to 0.1 Hz with amplitude of 32 mV .

2.5. Surface morphological studies

The alteration in the morphology of mild steel surface by the presence of inhibitor was studied by taking the images of metal specimens before and after immersion in corrosive medium with and without inhibitor for a period of 7 h using a Scanning Electron Microscope (JEOL JSM 6380 LA) with an accelerating voltage of 20 kV at a magnification of $\times 500$.

3. Results and discussion

3.1. Weight loss method

The weight loss method of corrosion monitoring being simple, accurate and reliable is considered as “gold standard” in corrosion testing. The mild steel corrosion in 0.5 M HCl in the absence and presence of the studied inhibitor HMATD was subjected to gravimetric weight loss technique for different immersion times 24 h, 48 h and 72 h. The corrosion rate (CR in $\text{mg cm}^{-2}\text{ h}^{-1}$) and inhibition efficiency (IE) were calculated using the equations [29]

$$CR = \frac{W}{Axt} \quad (1)$$

$$IE = \frac{W_0 - W_i}{W_0} \times 100 \quad (2)$$

where W is the weight loss before and after immersion in aggressive solution, W_0 and W_i represent weight loss in the absence and presence of inhibitor respectively, A the area of immersed metal coupon and t time in hours. The corrosion rate and inhibition efficiency obtained for various immersion times and various inhibitor concentrations are tabulated in Table 1. Variation of weight loss against concentration of HMATD for various immersion times in the absence and presence of HMATD is given in Fig. 4. The close examination of the table pinpoints the inverse relationship of inhibitor concentration with corrosion rate and direct relationship with inhibition efficiency. These trends suggest the role of adsorption and surface coverage which increases with increase in inhibitor concentration in corrosion inhibition. The increase in inhibition efficiency with inhibitor concentration is due to an increase in the number of molecules adsorbed on the metal surface at optimum concentration and more active sites of the metal are protected by the inhibitor molecules. [30].

3.2. Electrochemical measurements

3.2.1. Electrochemical impedance spectroscopy

3.2.1.1. Effect of inhibitor concentration. It is one of the important parameters for understanding the mechanism of corrosion, passivation, charge transfer etc. at the metal/electrolyte interface. It helps us to investigate the surface layer created by inhibitor on the metal. EIS studies on the mild steel corrosion in 0.5 M HCl solutions in the absence and presence of various concentrations of HMATD were performed at open circuit potential and the resulted

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