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New synthesised diamine derivatives as corrosion inhibitors of steel in 0.5 M H₂SO₄

A. Ouchrif^a, M. Zegmout^{b,d}, B. Hammouti^{a,*}, A. Dafali^a, M. Benkaddour^c, A. Ramdani^b, S. Elkadiri^d

^a Laboratoire de Chimie des Eaux et Corrosion, Faculté des Sciences, Universite Mohammed 1 er, Oujda 524, Morocco
 ^b Laboratoire de Chimie Organique Physique, Faculté des Sciences, Oujda, Morocco
 ^c Laboratoire d'Analyse et Caractérisation des Matériaux, Faculté des Sciences, Oujda, Morocco
 ^d Laboratoire de Chimie de l'Environnement et des Matériaux, Faculté des Sciences, Oujda, Morocco

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Abstract

The effect of addition of diamine derivatives on the corrosion of steel in $0.5\,\mathrm{M}$ H₂SO₄ solution is studied using weight loss measurements, electrochemical polarisation and impedance spectroscopy (EIS) methods. This study permits to follow the evolution of the inhibitive effect of diamine derivatives on steel in $0.5\,\mathrm{M}$ H₂SO₄. Polarisation measurements show that the diamines act as mixed inhibitors. The cathodic curves indicate that the reduction of proton at the steel surface happens within a pure activating mechanism. The inhibitors are adsorbed on the steel surface according to the Frumkin adsorption isotherm model. We note a good agreement between gravimetric, electrochemical polarisation and impedance spectroscopy methods.

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1. Introduction

Corrosive solutions in the acidic domain are more and more encountered in industry [1]. The importance of inhibitive protection in acidic solutions is increased by the fact that steels which are more susceptible to be attacked in aggressive media, are the commonly exposed metals in industrial environments. It is shown that the protective properties of diamine derivatives depend upon their ability to reduce corrosion rate and are enhanced at higher electron densities around the nitrogen atoms. Numerous works have been devoted to the corrosion inhibiting effect of aqueous soluble diamine on metallic materials [2].

The effect of organic nitrogen compounds on the corrosion behaviour of metallic materials in aggressive solutions has been well documented [3–16]. Their choice is based on their low-cost and stability as corrosion inhibitors for metallic ma-

terials in acidic media. The most synthesised compounds are the nitrogen-heterocyclic compounds, which are very known to be excellent complex or chelate forming substances with metals of transition series. Their adsorption is generally explained by the formation of an adherent film on the metal surface [17–19].

In the present work, we investigate the corrosion of steel in $0.5\,\mathrm{M}$ H₂SO₄ by some newly synthesised diamine compounds (Fig. 1), to point out any relation between the molecular structure of these compounds and their inhibitive action. Weight loss tests and two electrochemical techniques such as potentiodynamic polarisation and impedance measurements have been used to study the effect of addition of these compounds on the corrosion of steel in acid sulphuric solution.

2. Experimental

The diamine compounds are synthesised, purified and characterised by NMR, IR spectroscopies and element anal-

^{*} Corresponding author. Tel.: +212 6 74 47 48; fax: +212 6 74 47 49. E-mail address: hammouti@sciences.univ-oujda.ac.ma (B. Hammouti).

Diamine-1: 1,3- diaminopropane (M=74 g)

Diamine-2: ({3-[(cyanomethyl)amino]propyl}amino)acetonorile (M=152g)

Diamine-3: ({3-[(bis-cyanomethyl)amino]propyl}amino)acetonorile (M=191g)

Fig. 1. Molecular structures of diamine studied.

ysis before use. The molecular structures of diamines studied are shown in Fig. 1.

Prior to all measurements, the steel samples (0.09% P; 0.38% Si; 0.01% Al; 0.05% Mn; 0.21% C; 0.05% S and the remainder iron) are polished with different emery papers up to 1200 grade, washed thoroughly with bidistilled water, degreased and dried with acetone. The aggressive solution (0.5 M H₂SO₄) is prepared by dilution of analytical grade 98% H₂SO₄ with bidistilled water.

Gravimetric measurements are carried out in a double walled glass cell equipped with a thermostat-cooling condenser. The solution volume is $50 \, \text{cm}^3$. The steel specimens used have a rectangular form $(1 \, \text{cm} \times 1 \, \text{cm} \times 0.05 \, \text{cm})$.

Electrochemical experiments are recorded using a potentiostat model Voltalab PGZ 100. A platinum counter electrode and a saturated calomel electrode (SCE) are used. The working electrode (WE) in the form of a disc cut from steel, is embedded in polytetrafluoroethylene (PTFE).

The test solution is deaerated for 30 min in the cell with pure nitrogen. Gas pebbling is maintained through the experiments. The WE is then inserted and prepolarised at $-800 \,\mathrm{mV}$ (SCE) for 10 min in order to remove the oxide film from the electrode and E_{corr} is monitored until stationary. The scan rate is $20 \,\mathrm{mV} \,\mathrm{mn}^{-1}$.

The electrochemical impedance spectroscopy (EIS) measurements are carried out with a electrochemical system (Tacussel) which included a digital potentiostat model Voltalab PGZ 100 at $E_{\rm corr}$ after immersion in solution without bubbling. The circular surface of steel (1 cm²) exposed to the solution is used as working electrode. After the determination of steady-state current at a given potential, sine wave voltage (10 mV) peak to peak, at frequencies between 100 kHz and 10 MHz are superimposed on the rest potential. Computer programs automatically controlled the measurements performed at rest potentials after 30 min of

exposure. The impedance diagrams are given in the Nyquist representation.

3. Results and discussion

3.1. Weight loss tests

The effect of addition of diamine compounds tested at different concentrations on corrosion of steel in deaerated $0.5\,\mathrm{M}\,\mathrm{H}_2\mathrm{SO}_4$ solution is studied by weight loss at 298 K after 6 h of immersion. Inhibition efficiency (E_w , %) is calculated as follow:

$$E_{\rm w} (\%) = \left(1 - \frac{W_{\rm corr}}{W_{\rm corr}^{\rm o}}\right) 100$$

where $W_{\text{corr}}^{\text{o}}$ and W_{corr} are the corrosion rates of steel without and with diamine, respectively.

The results of weight loss of steel in 0.5 M H₂SO₄ with and without the addition of various concentrations of the diamine and derivatives are given in Table 1.

From gravimetric measurements, these results show for each compound tested, the steel corrosion rate values decrease when the concentration of diamine derivatives increases. The corrosion rate has a marked inhibitive effect, which attains 80 and 84% at 10^{-3} M for diamine-3 and diamine-2, respectively.

3.2. Electrochemical results

Current–potential characteristics resulting from cathodic and anodic polarisation curves of steel in 0.5 M H₂SO₄ in the absence and presence of diamine at various concentrations

Table 1
Gravimetric results of steel corrosion in 0.5 M H₂SO₄ without and with various concentrations of diamines at 298 K at 6 h

Concentration (M)		$W_{\rm corr} ({\rm mg/cm^2 h^{-1}})$	E _w (%)
Blank		1.780	_
Diamine-3	10^{-3}	0.290	84
	5×10^{-4}	0.351	80
	10^{-4}	0.549	69
	5×10^{-5}	0.719	59
	10^{-5}	1.221	31
	5×10^{-6}	1.389	22
Diamine-2	10^{-3}	0.357	80
	5×10^{-4}	0.450	75
	10^{-4}	0.769	57
	5×10^{-5}	0.908	49
	10^{-5}	1.258	29
	5×10^{-6}	1.329	25
Diamine-1	10^{-3}	0.481	73
	5×10^{-4}	0.862	69
	10^{-4}	1.154	57
	5×10^{-5}	1.176	40
	10^{-5}	1.252	30
	5×10^{-6}	1.340	25

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