International Journal of Science and Research (IJSR)
ISSN (Online): 2319-7064
Impact Factor (2012): 3.358

Some Triazolic Compounds as Corrosion Inhibitors for Copper in Sulphuric Acid

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Abstract: ***Corrosion inhibition of copper in 1.0 M H_2SO_4 was investigated in the absence and presence of different concentrations of triazole derivatives namely, 1,2,4-triazole (TR), 3-amino-1,2,4-triazole (ATA) and 2-4-diamino-1,2,4-triazole (DTA). Weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) measurements were employed. Impedance measurements showed that the charge transfer resistance increased and double layer capacitance decreased with increase in the presence of these inhibitors. Potentiodynamic polarization reveal that compound DTA is the best inhibitor and the inhibition efficiency (IE%) follows the sequence: DTA > ATA > TR. Obvious correlations were found between corrosion inhibition efficiency and some quantum chemical parameters such as energy of highest occupied molecular orbital (HOMO), energy of lowest unoccupied molecular orbital (LUMO), HOMO–LUMO energy gap and electronic density etc. Calculated results indicated that the difference in inhibition efficiencies between these compounds can be clearly explained in terms of frontier molecular orbital theory. The inhibitor performance depends mainly on the type of function groups substituted on triazole ring.***

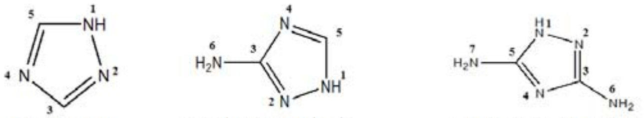
Keywords: Corrosion, Inhibition, Copper, Triazole

1. Introduction

Copper plays a significant role in many cultures. It is a major industrial metal by its electrical and thermal properties [1]. Because of the general aggressiveness of acid solutions, the use of inhibitors to control the destructive attack of acid environment was found to have widespread applications in many industries. Using inhibitors is an effective method to reduce corrosion rate [1]. An acid wash process is widely used in many industries in order to cleaning and descaling of metallic substrates [2]. In acid pickling process, hydrochloric acid, phosphoric acid and sulfuric acid are used due to their special chemical properties [3]. By considering the fact that mentioned mineral acids are aggressive solution for metallic substrates, using corrosion inhibitors in acidic bath is one of the most practical methods for preventing the corrosion of metals. As a result, corrosion inhibitors for acidic media have attracted increasing attention due to their extended applications [3–5]. The use of organic compounds containing oxygen, sulfur and nitrogen has been studied in some details to reduce corrosive attack on copper in acid medium [6–14]. In recent years, the quantum chemistry computing method has become an effective way to study the correlation of the molecular structure and its inhibition properties and much achievement was reached [15–20]. The present work is an extension of earlier works on the influence in a series of the triazoles derivative (1,2,4-triazole (TA), 3-amino-1,2,4-triazole (ATA) and 2-4-diamino-1,2,4-triazole (DTA) on the inhibition of corrosion of copper in 1M H2SO4. The inhibition aimed performance is evaluated by weight loss, Tafel polarization and electrochemical impedance spectroscopy (EIS). This study is also to show the formation of copper-ATA and copper-DTA complexes and to find a correlation between the highest occupied molecular orbital energy EHOMO and inhibition efficiencies.

2. Experimental

Triazolic compounds tested were 1, 2, 4-triazole (TR), 3-amino-1,2,4-triazole (ATA) and 2-4-diamino-1,2,4-triazole (DTA). They are Aldrich commercial products (> 99 % purity). Their molecular structures are shown in Figure 1.



1-2-4Triazole 3-Amino-1-2-4-Triazole 3-5-Diamino-1-2-4-Triazole

Figure 1: chemical structure of triazolic compounds (TR, ATA, DTA)

The aggressive solution (1M H_2SO_4) was prepared by dilution of Analytical Grad 98% H_2SO_4 with bidistilled water. The weight loss measurements were carried out in a 100 mL capacity glass beaker placed in a thermostat water bath. The solution volume was 80 mL. The used copper coupons (99.99 % of purity) had a rectangular form (length = 2.5 cm, width = 2 cm, thickness = 0.05 cm). The coupons were weighed and suspended in 100 mL of an aerated 1.0 M H_2SO_4 solution containing Triazolic derivatives at the desired concentrations exposure time varying from 4 to 168 h at (298 ± 1) °C. At the end of the tests, the coupons were taken out, washed with bi-distilled water, degreased with acetone, washed again with bi-distilled water, dried, and then weighed using an analytical balance (precision: ±0.1 mg). Three measurements were performed in each case and the mean value of the weight loss has been reported.

For the electrochemical studies, the electrode was provided in rod form and was embedded in epoxy resin in a Teflon holder with electrical contact being achieved by means of a copper wire threaded into the base of the metal sample. The surface of the specimen was 0.35 cm^2. Prior to each test, the exposed surfaces were abraded successively with different emery paper up 1200 grade, washed thoroughly with acetone and rinsed with double distilled water.

The current–voltage characteristics are recorded with a potentiostat PGZ100 piloted by Voltamaster soft-ware. The scan rate is 60 mV/min and the potential is ranged from cathodic to anodic potentials. Before recording each curve, the working electrode is maintained with its free potential of corrosion E_{corr} for 30 min. We used for all electrochemical tests a cell with three electrodes and double wall thermostats (Tacussel Standard CEC/TH). Saturated calomel (SCE) and platinum electrodes are used as reference and auxiliary electrodes, respectively.

The tests were carried out in a temperature range from 298 to 323 K. The electrochemical impedance spectroscopy (EIS) measurements are realised with the electrochemical system (Tacussel), which included a digital potentiostat model Voltalab PGZ100 computer at E_{corr} after 30 min immersion in solution. After the determination of steady-state current at a corrosion 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. The impedance diagrams are given in the Nyquist representation. Experiments are repeated three times to ensure the reproducibility.

3. Computational Details

The correlation between theoretically calculated properties and experimentally determined inhibition efficiencies has been studied successfully for uniform corrosion [21–23]. To support experimental data, theoretical calculations were conducted in order to provide molecular-level understanding of the observed experimental behavior. The major driving force of quantum chemical research is to understand and explain the functions of these triazolic compounds derivatives in molecular terms. Several theoretical parameters includes the electronic properties of inhibitors, effects of the frontier molecular orbital energies, the differences between lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) energies ($E_{HOMO} - E_{LUMO}$), electronic charges on reactive centers, dipole moments and conformation of molecules have been investigated by semi-empirical methods.

In this work, calculations are performed by using restricted Hartree-Fock level using Austin Model 1 (AM1), Modified Neglect of Differential Overlap (MNDO) and Parametric Model 3 (PM3) implanted in MOPAC 6.0 program.

4. Results and Discussions

4.1 Weight Loss Measurements

The gravimetric method (weight loss) is probably the most widely used method of inhibition assessment. The simplicity and reliability of the measurement offered by the weight loss method is such that the technique forms the baseline method of measurement in many corrosion monitoring programmers' [24, 25]. The corrosion rate (w) was calculated from the following equation [26]:

$$W = \frac{\Delta m}{S.t} \tag{1}$$

where Δm is the average weight loss of copper sheets, S the total area of one copper specimen, and **t** is the immersion time. With the estimated corrosion rate, the inhibition efficiency E_W (%) was calculated as follows:

$$Ew(\%) = \frac{Wcorr - W'corr}{Wcorr} \times 100 \tag{2}$$

where w_{corr} and w'_{corr} are the corrosion rates of the copper coupons in the absence and presence of inhibitor, respectively. The evolution of weight loss data of copper in H_2SO_4 with time immersion in the absence and presence of 10^{-3} M of inhibitor is shown in figure 2.

According to this data, it is clear that the addition of triazolic compounds reduces the corrosion rate of copper in sulphuric acid solution. The lower corrosion rate is obtained in presence of DTA after 168 h exposure time. The inhibition efficiency (E_G%) was calculated using Eq. (2). The average corrosion rates, expressed in $mg.cm^{-2}.h^{-1}$, are given in Table 1. The results show that all triazole derivatives used inhibit the corrosion of copper in 1M H_2SO_4 solutions. The inhibition efficiency increases in the sequence:

AT < ATA < DTA. DTA is better inhibitor than both ATA and TA.

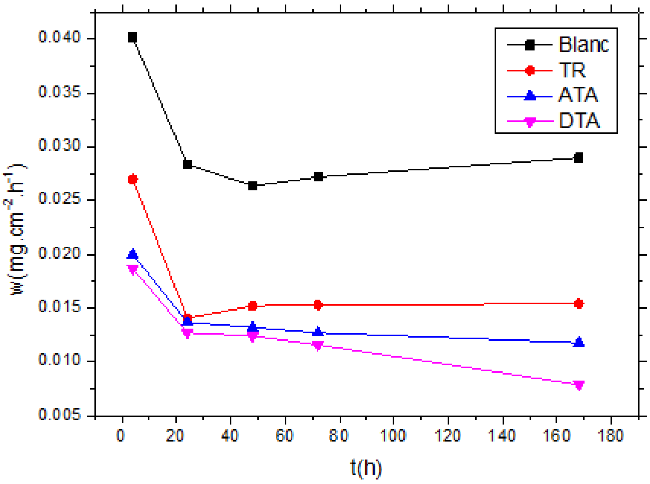


Figure 2: Corrosion rate of copper in H_2SO_4 as a function of time without and with addition of the inhibitors at 10^{-3}M

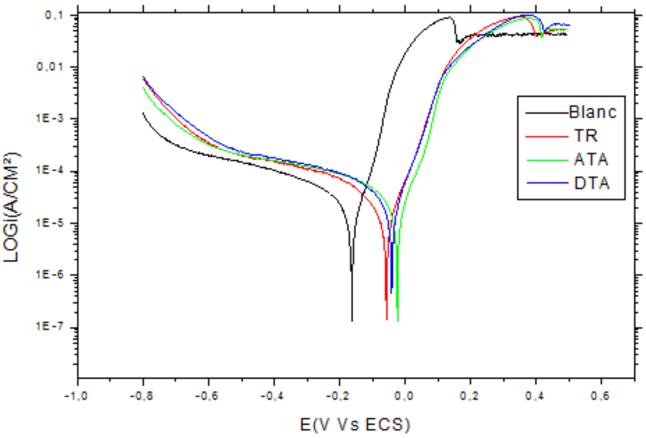


Figure 3: Polarization curve of copper in medium H_2SO_4 in the absence and presence of 10^{-3} M of Triazole derivatives

Table 1: Inhibitory efficiencies of Triazolic compounds at 10^{-3} M as a function of immersion time

E_w (%)					
Time immersion	4 h	24 h	48 h	72 h	168 h
H_2SO_4 1M	-	-	-	-	-
H_2SO_4 1M + AT	**33**	**51**	**42**	**43**	**47**
H_2SO_4 1M + ATA	**50**	**52**	**50**	**53**	**59**
H_2SO_4 1M + DTA	**54**	**55**	**53**	**57**	**73**

4.2 Tafel Polarization Measurements

The polarization curves of copper in 1M H_2SO_4 in absence and presence of triazolic compounds are presented in Figure 3. We note that all triazole derivatives show relatively similar behavior in polarization testing. Figure 3 shows that these compounds do not change the profile of the anodic and cathodic curves, indicating that they block the reaction sites of copper without affecting the anodic and cathodic reaction mechanisms.

In general, copper can hardly be corroded in the deoxygenated dilute sulfuric acid [27], as copper cannot displace hydrogen from acid solutions according to theories of chemical thermodynamics. However, this situation will change in aerated sulfuric acid. Dissolved oxygen may be reduced on copper surface and this will enable some corrosion to take place [28]. It is a good approximation to ignore the hydrogen evolution reaction and only consider oxygen reduction in the aerated sulfuric acid solutions at potentials near the corrosion potential, according to Smyrl [29].
Cathodic reduction of oxygen can be expressed either by a direct four electrons transfer as shown by equation.

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \qquad (1)$$

or by two consecutive $2e^-$ steps involving a reduction to hydrogen peroxide first

$$O_2 + 2H^+ + 2e^- \rightarrow H_2 \qquad (2)$$

followed by a further reduction [30]

$$H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O \qquad (3)$$

Dissolution of copper in sulfuric acid is described by the following two consecutive steps:

$$Cu \rightarrow Cu^+_{ads} + e- \qquad (4)$$

$$Cu^+_{ads} \rightarrow Cu^{2+} + e^- \qquad (5)$$

Where Cu^+_{ads} is an adsorbed species at the copper surface and does not diffuse into the bulk solution [31]. The measured parameters such as corrosion potential (E_{corr}), corrosion current density (I_{corr}) and inhibition efficiency (E_I%) are listed in Table 3. Equation (5) determines the inhibition efficiency:

$$EI(\%) = \frac{Icorr - I'corr}{Icorr} \times 100 \qquad (5)$$

where I_{corr} and I'_{corr} are corrosion current densities without and with inhibitors respectively, as determined by extrapolation of anodic Tafel lines to corrosion potential [32]. Thus, the presence of triazolic compounds at 10^{-3} M leads to decrease in the values of I_{corr}, which was particularly significant in the case of DTA. Thus, the presence of triazolic compounds at 10^{-3} M leads to decrease in the values of I_{corr}, which was particularly significant in the case of DTA.

Table 2: Electrochemical parameters and inhibition efficiency of copper in 1M H_2SO_4 medium in the absence and presence of 10^{-3}M of Triazole compounds

Medium	E_{corr} (mV/ECS)	ba (mV/dec)	I_{corr} (μA/cm^2)	E_I (%)
Blank	-131	65	33	_
Triazole	-59	66	11	66
Amino-triazole	-24	64	10	71
Diamino-triazole	-42	68	09	72

4.3 EIS Measurements

The corrosion inhibition of copper in acidic solution in the presence and absence of inhibitors AT, ATA, DTA, at 10^{-3}M, is also investigated by the electrochemical impedance spectroscopy (EIS) at 298 K after 30 min of immersion. These diagrams were obtained at open circuit potential in a frequency range between 100 kHz to 10 mHz and an amplitude of 10 mV. The Nyquist plots for the copper plates in corrosive media (1 M H_2SO_4) in the absence and presence of different triazolic compounds are shown in Figure 4. This figure shows that the Nyquist plots of copper in acidic medium consist of distorted semicircles. The increase in the size of the semicircle in presence of the inhibitors indicates that a barrier gradually forms on the copper surface. The charge-transfer resistance (R_t) values are calculated from the difference in impedance at lower and higher frequencies [25]. To obtain the double layer capacitance (C_{dl}) the frequency at which the imaginary component of the impedance is maximal ($-Z_{max}$) is found as represented in equation:

$$Cdl = \frac{1}{\omega.Rt} \qquad (6)$$

where $\omega = 2\pi f_{max}$, is frequency in rad.s^{-1} and f_{max} is the frequency in Hz. The percentage of inhibition efficiency (E %) was calculated from the impedance measurements using the relation:

$$E_{Rt}(\%) = \frac{R_t' - R_t}{R_t'} \times 100 \qquad (7)$$

where R_t' and R_t are the charge transfer resistance values with and without inhibitor, respectively. Table 4 reported different values of electrochemical parameter deduced from EIS study. Increasing R_t values in presence of triazolic compounds, suggesting decrease of the corrosion rate since the R_t value, is a measure of electron transfer across the surface, and inversely proportional to the corrosion rate. The decrease in the C_{dl} values could be attributed to the adsorption of the chemical constituents of inhibitors at the metal surface [33]. The data shown in table (3) indicate that addition of triazolic compounds leads to increase of the charge transfer resistance which is associated with a decrease of double layer capacitance. E_{Rt} varied in the sequence: AT < ATA < DTA. The inhibition efficiency obtained from EIS measurement are close to those deduced from polarization and gravimetric methods.

The corrosion inhibition can be attributed to the adsorption of triazoles molecules at copper acid solution interface. Adsorption of triazole derivatives can be explained on the basis that adsorption of these compound are mainly via the nitrogen atoms in the triazole ring, in addition to the availability of π electrons (by resonance structures) in the aromatic system. In case of AT and DAT, the presence of amino group enhances its adsorption more than TR itself.

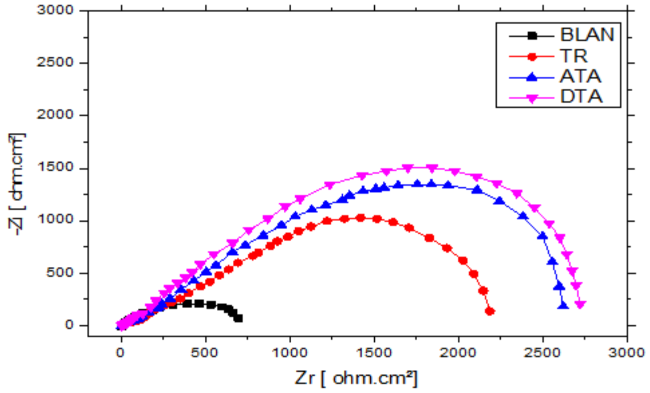


Figure 4: Impedance diagram of copper in 1M H_2SO_4 without and with the addition of triazoles at 10^{-3}M

Table 3: Impedance parameters for copper in 1M H_2SO_4 with and without addition of triazolic inhibitors at 10^{-3} M, and the corresponding inhibition efficiency

Medium	R_t ($\Omega.cm^2$)	f_m (Hz)	C_{dl} ($\mu F.cm^{-2}$)	E_{Rt} (%)
Blank	749	2.13	99.95	-
TR	2200	0.75	96.50	66
ATA	2623	0.67	90.60	71
DTA	2750	0.64	89.08	73

4.4 Theorical Correlation

More information may also be obtained from the calculated quantum chemical indices such as E_{HOMO}, E_{LUMO} and energy gap $\Delta E = E_{HOMO} - E_{LUMO}$ and ΔH_f obtained by semi-empirical AM1, MNDO, and PM3 methods. HOMO is the highest occupied molecular orbital and LUMO is the lowest unoccupied molecular orbital. Values of energy of LUMO and HOMO, ΔE, the formation heat ΔH_f and inhibition efficiencies, are given in Table 4. E_{HOMO} is often associated with the electron donating ability of the molecule. High values of E_{HOMO} indicate a tendency of the molecule to donate electrons to appropriate acceptor molecules with low energy of empty atomic orbital. Therefore, the energy of the lowest unoccupied molecular orbital indicates the ability of the molecule to accept electron. Also, the less negative HOMO and the smaller energy gap are reflected in stronger chemisorption bond and perhaps greater inhibitor efficiency. Examination of Table 4 reveals that ΔE obtained by the three methods decreases from TR to DTA. This result indicates that electron transfer between molecular orbital HOMO and LUMO is facilitated in the case of DTA which presents the maximum of inhibition efficiency. The charge transfer from inhibitor molecule takes place during its adsorption on the metal surface. The increases of EHOMO from TR, ATA to DTA facilitate probably the adsorption and thereafter the inhibition by supporting the transport process through the adsorbed layer [34]. The distribution of charge calculated by the three methods on the molecular structure is presented in Table 6. The negative charge on nitrogen atoms indicates that the triazolic inhibitors are adsorbed among the N atoms on the copper surface. It is clear that the substitution of two hydrogen atom in TR by one NH_2 in ATA is 71%. The second substitution of hydrogen by another NH_2 in DTA significantly reinforces the inhibitory efficiency of DTA to 73%. We may then introduce an intramolecular synergistic effect between all active sites of DTA molecule.

Correlation between the mean efficiency (E_m%) of the tested inhibitors calculated by the three methods and E_{HOMO} is determinate by equation (8):

$$E_m(\%) = \frac{E_W\% + E_I\% + E_{Rt}\%}{3} \qquad (8)$$

The ionization potential (I) and the electron affinity (A) are calculated from the Koopmans' theorem which establishes a relation between the energies of the HOMO and the LUMO and I and A, respectively, [35]. The values of I and A obtained were considered for the calculation of the electronegativity χ and global hardness η in each of inhibitor [36]. The fraction of electrons ΔN transferred from the inhibitor molecule to the metallic atom was calculated according to Pearson's method [37], the idea behind this is that in the reaction of two systems with different electronegativities (as a metallic surface and an inhibitor molecule), the following mechanism will take place: the electron flow from the molecule with the low electronegativity towards that of a higher value will occur, until the chemical potentials equalises. In order to calculate the fraction of electrons transferred, a theoretical value for the electronegativity of bulk copper according to Pauling, χ Cu = 1,9 eV, was used and a global hardness of ηCu = 0, by assuming that for a metallic bulk I = A [35] because they are softer than the neutral metallic atoms. For the calculation, the following formula, equation (9), was used [37]:

$$\Delta N = \frac{\chi Cu - \chi inh}{2(\eta Cu + \eta inh)} \qquad (9)$$

In the same way, it is possible to observe that the DTA molecule has a lower value of global hardness and electronegativity, Thus, the fraction of transferred electrons is largest for the DTA confirming that DTA is the best inhibitor, The AM1 method permits also the calculation of enthalpy of formation ΔH_f which is lower for DTA (Table 4), these values can be compared with those reported in previous works conducted on different molecules [38]. Concluding that the calculated fraction of electrons transferred for the systems under study in this work is considerably significant.

Table 4: AM1, MNDO and PM3 Theoretical Parameters calculated for the molecules tested

Inhibitors	ΔH_f (kcal/mol)	E_{HOMO} (eV)	E_{LUMO} (eV)	$E_{HOMO} - E_{LUMO}$ (eV)
AM1 TR ATA DTA	76.973 78.929 80.457	-10.270 -9.025 - 8.818	0.542 0.671 0.657	10.812 9.697 9.475
MNDO TR ATA DTA	43.743 40.938 38.299	-10.310 -9.499 - 9.392	0.479 0.477 -0.406	10.790 9.978 9.798
PM3 TR ATA DTA	51.827 49.040 46.497	-10.396 -8.929 -8.819	0.104 0.217 0.204	10,500 9.147 9.024

Table 5: Atomic charges of inhibitors studied calculated by AM1, MNDO and PM3 methods

Inhibitors	Nette Charge	
	N1 N2 C3 N4 C5 N6 N7	ΣQi
AM1 TA ATA DTA	-0.197 -0.160 -0.091 -0.172 -0.156 -0.199 -0.137 -0.011 -0.185 -0.068 -0.273 -0.199 -0.141 0.001 -0.218 0.058 -0.272 -0.281	-0,802 -0,825 -0,971
MNDO TA ATA DTA	-0.198 -0.140 -0.003 -0.251 0.075 -0.199 -0.162 0.077 -0.263 0.092 -0.192 -0.183 -0.167 0.088 -0.282 0.156 -0.194 -0.210	-0,547 -0,349 -0,563
PM3 TA ATA DTA	-0.185 -0.170 -0.076 -0.165 -0.075 -0.189 -0.136 -0.082 -0.193 -0.063 -0.141 -0.189 -0.180 0.063 -0.225 -0.055 -0.220 - 0.231	-0,526 -0,631 -0,606

Table 6: Parameters electronic of molecules studied

Molecule	I=- E_{HOMO}	A=- E_{LUMO}	$\chi = (A+I)/2$	$\eta = (I-A)/2$	ΔN
TR	10.27 0	- 0.542	4.862	5.406	- 0.274
ATA	9.025	- 0.657	4.182	4.841	- 0.236
DTA	8.818	- 0.671	4.074	4.745	- 0.229

5. Conclusion

We have studied the inhibiting effect of triazolic compounds on the copper corrosion in 1M H_2SO_4 by using various methods. From the above results and discussion, the following conclusions are drawn:

- Triazolic derivatives were found to be effective inhibitors for copper corrosion in 1M H_2SO_4 solutions. Corrosion inhibition efficiency depends on the chemical structure of inhibitors. At the concentrations studied, inhibition performance follows the sequence AT < ATA < DTA.
- Double layer capacitances decreases with respect to the blank solution when these inhibitors are added, this fact may be explained on the basis of adsorption of these inhibitors on the copper surface.
- In determining the corrosion rates, electrochemical studies and weight loss measurements gave similar results.
- The quantum mechanical approach may well be able to foretell molecular structures that are better for corrosion inhibition.

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