Int. J. Electrochem. Sci., 8 **(2013)** 6513 - 6523

International Journal of
ELECTROCHEMICAL
SCIENCE
www.electrochemsci.org

Inhibition of the Corrosion of Copper in a Sodium Chloride Solution Using 4-((4-dodecylphenylimino)methyl)phenol

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Received: 3 March 2013 / *Accepted:* 23 March 2013 / *Published:* 1 May 2013

A new kind of Schiff base, 4-((4-dodecylphenylimino)methyl)phenol (DPMP), is synthesized. Its structure is characterized by Fourier-transform infrared (FT-IR) and ^{1}H nuclear magnetic resonance. Self-assembled films of DPMP are prepared on a copper surface. Electrochemical impedance spectroscopy (EIS) and polarization curve analyses are performed to measure the protection ability against copper corrosion in a 0.5 mol/L NaCl solution. FT-IR spectroscopy and scanning electron microscopy are used for the surface topography measurements. The results indicate that DPMP could self-assemble on the copper surface. The inhibition efficiency (*IE*) increases with increasing DPMP solution concentration. The maximum *IE* in 0.01 mol/L DPMP-ethanol solution after 2 h of self-assembly is 99.87%.

Keywords: Schiff base; Copper corrosion; Electrochemical Impedance spectroscopy; Polarization curve; Inhibition efficiency

1. INTRODUCTION

The technology of self-assembled films involves the spontaneous formation of active molecules or atoms into ordered thin films on a substrate via chemical bonding. A chemical reaction spontaneously occurs when the substrate is immersed in a solution containing active molecules or atoms. The self-assembly technology is convenient to operate, and the experimental equipment is simple. The molecules on the substrate have ordered arrangements and high thermodynamic stabilities [1, 2]. The self-assembly technology is widely used in mesoporous materials, nanoparticles, biosensors, and corrosion inhibition [3-6]. Ultrathin self-assembled films have recently been developed for use in anticorrosion. These films can protect metals from corrosion because of their densely packed

and stable structures [7]. Compared with traditional corrosion inhibition technologies, ultrathin films have higher inhibition efficiencies (*IE*s) and consume less raw materials.

Numerous compounds subjected to the self-assembly technology have been used as corrosion inhibitors. Alkyl thiols [8-10], fatty acids and their derivatives [11, 12], phosphates, imidazole derivatives [13], as well as Schiff bases have all been used as inhibitors to prevent metal corrosion.

Schiff bases could be easily synthesized at a low cost. Apart from their use as fungicides, their nontoxicity enables other extensive applications. As a new group of corrosion inhibitors, Schiff bases have been recently employed in studies on copper surface corrosion. Some Schiff bases could inhibit metal corrosion, such as N,N'-ethylen-bis (salicylidenimine) [14], benzylidene-phyrimidine-2-yl-amine [15], 2-((5-mercapto-1,3,4-thiadiazol-2-ylimino)methyl)phenol [16], 4,4'-bis(3-carboxaldehyde thiophene) diphenyl diimino ether [17], and 2-(2-hydroxyphenyl)-2,5-diaza-4,6-dimethyl-8-hydroxy-1,5,7-nonatriene [18]. Although the self-assembled films of these compounds are reported to have high *IE* values, the films are not complete. These compounds mainly contain conjugated structures with short alkyl chains. The ions or oxygen molecules in the solutions of such compounds could easily make contact with metals and cause corrosion.

The long alkyl chains of thiols prevent metal contact with a solution, forming a hydrophobic layer on the metal surface. However, the films can easily disintegrate, indicating a need for increasing the number active groups. Therefore, new Schiff bases containing long alkyl chains and more than one active group need to be designed and synthesized. Importantly, such compounds must be able to improve the corrosion resistance of metals. There is no report in literature on the use of long-chain-alkane Schiff bases in the field of metal corrosion inhibition.

In the current work, a new Schiff base, 4-((4-dodecylphenylimino)methyl)phenol (DPMP), is designed and synthesized. Its hydroxyl and —C=N— functional groups provide lone pair electrons that enable its stable adsorption on a copper surface. In high concentrations, multilayer DPMP films that form on the copper surface are very stable. When DPMP is compactly and orderly arranged on the copper surface, its hydrophobic long alkyl chain act as a hydrophobic layer, could effectively block chlorine ions from NaCl solution from making contact with the copper surface. Consequently, metal corrosion is inhibited. Fourier-transform infrared (FT-IR) and scanning electron microscopy (SEM) are used to characterize the self-assembled films on the copper surface. To study the effect of the DPMP concentration on copper corrosion in 0.5 mol/L NaCl solution, electrochemical impedance and polarization curve measurements are conducted. The activation parameters influencing the inhibition behavior of the compound are calculated, and the factors affecting the self-assembly are discussed.

2. MATERIAL AND METHODS

2.1. Chemical materials

The reagents used in this experiment were all analytical grade chemicals. 4-Hydroxybenzaldehyde (PHBA), 4-dodecylaniline and other chemicals used in the experiment were

commercially obtained and used without further purification. The 0.5mol/L NaCl solution was prepared by dissolving NaCl in ultrapure water.

2.2. Preparation of Schiff base

DPMP was synthesized from 4-hydroxybenzaldehyde and 4-dodecylaniline. 4-Dodecylaniline (2.6121 g, 9.99×10^{-3} mol) and 4-hydroxybenzaldehyde (1.2221 g, 1.00×10^{-2}mol.) were dissolved in 50mL ethanol. The mixture reacted for 3 h in condensing circumfluence equipment. Continuous stirring was performed under the temperature of 80°C for 3 h. After that, the reaction mixture was heated to half the volume, cooled down to crystallize overnight. The recrystallization product was dried at 75°C. The melting point of the compound was obtained via a digital melting point apparatus (WRS-1B).

For the synthetic compound: light orange; m.p. 144.4 °C. FT-IR (ν, cm^{-1}): 3435.7 (OH), 3028.4 (Ar—H), 2958.2, 2919.1, 2850.2 (CH_{st}), 1606.1 (C=N) [19], 1575.7-1443.5 (C=C), and 840.7 (orienting group). ^{1}H NMR (δ, ppm) ($DCCl_3$): 8.4 (s, OH); 7.7, 7.2, 6.8 (m, 4H, ArH); 2.6 (s, CH); 1.6 (t, CH_2); 1.3 (m, CH_2); 0.9 (t, CH_3).

The compound was dissolved in anhydrous ethanol to the desired concentration from 10^{-5} mol/L to the maximum concentration of 10^{-2} mol/L.

2.3. Preparation of electrodes and self-assembled films

The working electrodes were prepared from pure copper 4.12mm in diameter and 2~3cm long. Pure copper sheets (about $1cm^2$) were used as the substrate for SEM, AFM, and FT-IR experiments. After deposits were removed by using acids, the surface of the copper rods was packed with epoxide resin leaving only the cross-section contact with the electrolyte solution. The other end of the electrode was connected with Cu-wire.

The copper electrode and the sheet were first ground with silicon carbide abrasive papers from 800 to 2500 grain, polished with alumina paste (0.05 μm) to achieve a mirror finish, rinsed with ultrapure water, degreased with ethanol, dried in a stream of nitrogen, and then etched in 7 mol/L HNO_3 for 15 s to obtain a fresh copper surface. The electrode was then rinsed with ultrapure water and absolute ethanol, dried, and immersed in DPMP-ethanol solution immediately. Self-assembled copper materials were rinsed with ultrapure water, dried with nitrogen for further measurements.

2.4. Electrochemical measurements

Electrochemical experiments were performed in a three-electrode cell. Copper electrode, platinum slice (about $1cm^2$) and saturated calomel electrode (SCE) were used as the working electrode, auxiliary electrode and reference electrode, respectively. Potentials reported were all referred to the SCE. The electrochemical experiments were carried out using the electrochemical measurement system Potentiostat Model 263A workstation (PerkinElmer) with a lock-in amplifier (Model 5210;

10mHz ~100kHz; PerkinElmer). Data were collected, and figures were plotted using the software Powersuit.

Electrochemical impedance spectroscopy (EIS) measurements were carried out at the corrosion potential. The amplitude of the sinusoidal perturbation signal was 10 mV at the open circuit potential (OCP) in the frequency range 100 kHz to 10 mHz, with 30 points per decade. The ZSimpWin software was used to analyse the impedance data. The polarization curves were carried out at a scan rate of 0.1660 mV/s constructed within the potential range of ±0.25 V versus OCP.

2.5. FT-IR spectroscopy

A Nicolet 5700 Fourier transform infrared spectrometer (Thermo Electron) was used to test the FT-IR spectra of DPMP sample and modified copper sheet. 1mg of sample was added in about 200mg of IR spectroscopic grade KBr. Modified Copper sheet (self-assembly for 2h in 0.01mol/L DPMP-ethanol solution) was measured with reflectance spectra. FT-IR spectra was obtained in the range of 400~4000cm^{-1} at room temperature.

2.6. SEM

Four copper sheets prepared as described above (Section 2.3) were used as blank, blank-corrosion, self-assembly and self-assembly-corrosion samples, respectively. The corrosion was carried out in 0.5 mol/L NaCl solution for 1 h, and the self-assembly was in a 0.01 mol/L DPMP solution for 2 h. The copper sheets' morphology was investigated using a scanning electron microscope (FEI Sirion 200).

3. RESULTS AND DISCUSSION

3.1. Characterization of the synthetic compound

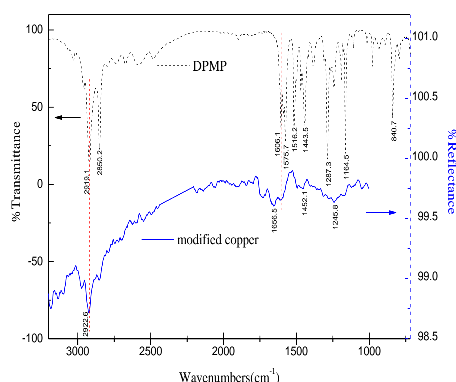


Figure 1. FT-IR of 4-((4-dodecylphenylimino) methyl) phenol and the copper sheet modified with 4-((4-dodecylphenylimino) methyl) phenol

The FT-IR spectra of DPMP and the modified copper sheet are shown in Fig. 1. Modified copper has the main peaks of the compound (ν, cm^{-1}): 2922.6 (CH_{st}), 1656.5 (C=N), and 1452.1 (C=C). These peaks confirm the presence of DPMP molecules on the copper surface. The acyl group wavenumber moved from 1601.1 cm^{-1} to 1656.5 cm^{-1}. It indicates that the lone pair electrons in N atom chelated with Cu. Hence, DPMP has been successfully adsorbed on the copper surface.

3.2. SEM

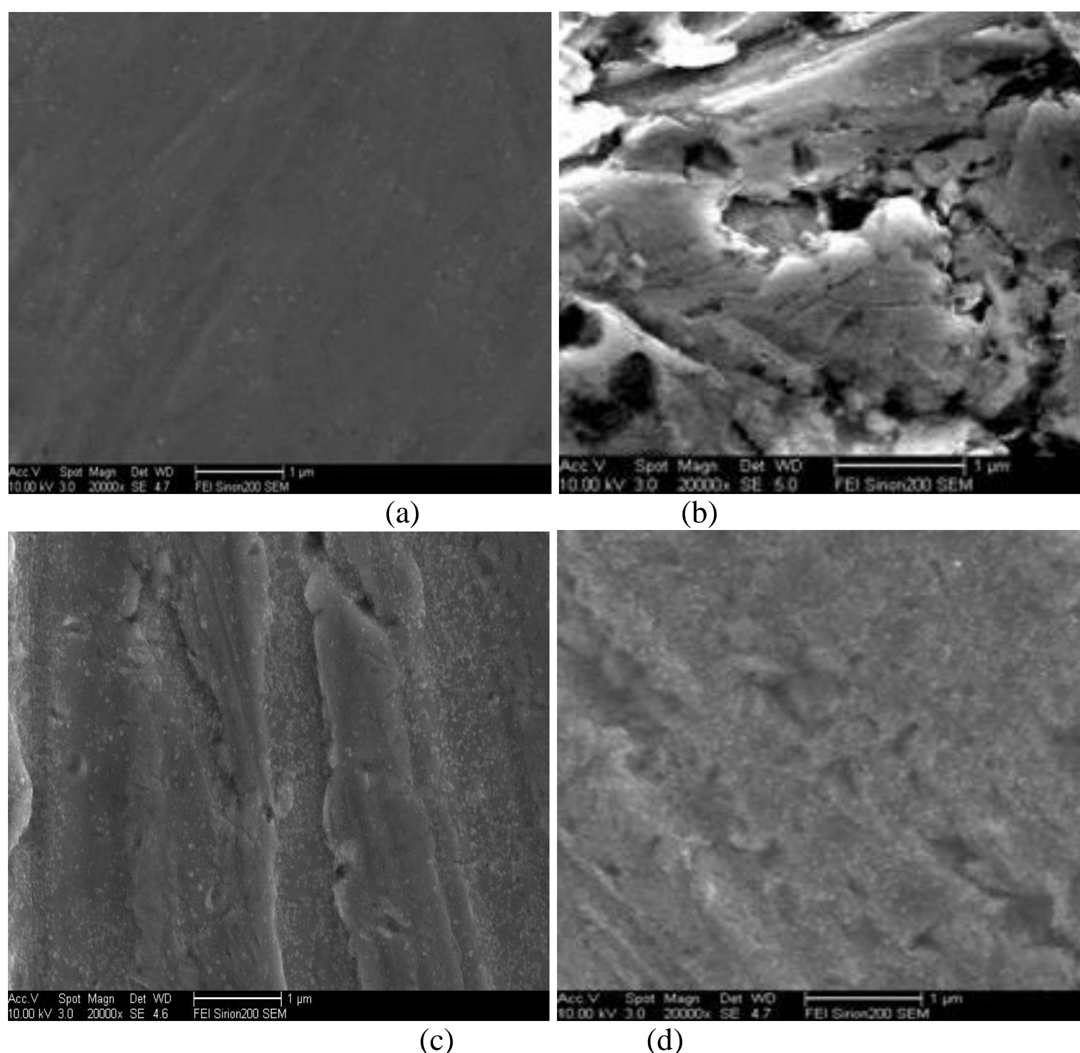


Figure 2. SEM of copper surfaces: (a) blank copper sheet; (b) blank copper sheet immersed in 0.5 mol/L NaCl solution for 1 h; (c) copper sheet self-assembled in DPMP-ethanol solution for 2 h; (d) copper sheet immersed in DPMP-ethanol solution for 2 h then immersed in 0.5 mol/L NaCl for 1 h

The SEM images are shown in Fig. 2. Fig. 2a shows the SEM image of the blank copper sheet surface. Fig. 2b shows the SEM image of the blank copper sheet immersed in 0.5mol/L NaCl solution for 1h, wherein serious corrosion can be observed. Fig. 2c shows the SEM image of the self-assembled copper sheet immersed in a 0.01 mol/L DPMP-ethanol solution for 2 h. The copper surface in Fig. 2c is irregular compared with that in Fig. 2a. At the same amplified time, the difference between these

surfaces illustrates the adsorption of DPMP molecules, i.e., self-assembled films, on the copper surface. Fig. 2d shows the SEM image of the copper sheet initially immersed in DPMP-ethanol solution for 2 h, and then in 0.5 mol/L NaCl solution for 1 h. The surface has no difference with that in Fig. 2c, indicating that the self-assembled films significantly hinders copper corrosion in 0.5 mol/L NaCl solution.

3.3. Polarization measurements

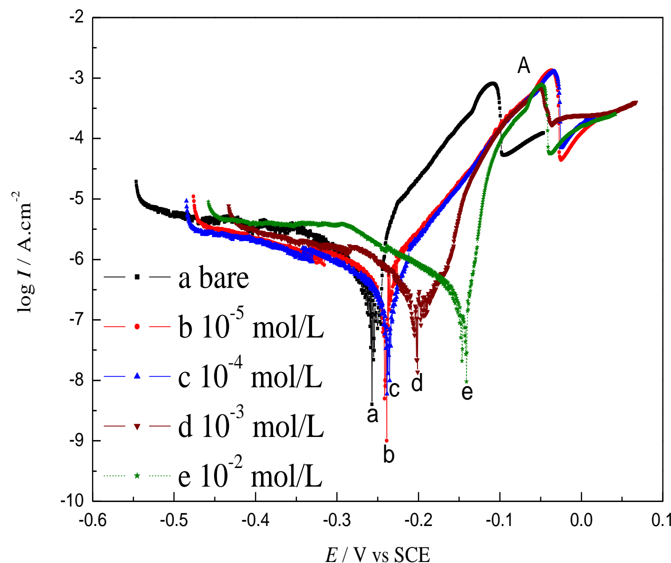


Figure 3. Polarization curves of copper electrodes self-assembled in different concentrations of DPMP-ethanol solution for 2 h at room temperature: (a) bare copper; (b) 10-5 mol/L solution; (c) 10-4 mol/L solution; (d) 10-3 mol/L solution; (e) 10-2 mol/L solution. Test environment, 0.5 mol/L NaCl solution

Table 1. Tafel polarization parameter values for the corrosion of the copper electrode with or without self-assembled films at different DPMP concentrations in 0.5 mol/L NaCl solution

Concentration (mol/L)	E_{corr} (V)	j_{corr} (μA•cm^{-2})	*IE* (%)
bare	-0.257	8.13	
10^{-5}	-0.239	2.24	72.46
10^{-4}	-0.238	0.407	94.99
10^{-3}	-0.201	0.273	96.64
10^{-2}	-0.141	0.0458	99.44

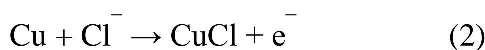
Fig. 3 presents the polarization curves of both the anode and cathode of a copper electrode in 0.5 mol/L NaCl solution self-assembled for 2 h in DPMP-ethanols solutions with different concentrations. It shows typical polarization curves for copper in 0.5mol/L NaCl solution with and without DPMP. The anodic curve for the copper electrode in a 0.5 mol/L HCl blank solution exhibits

two limiting current plateaus. This indicates the role of a diffusion-limiting step, probably both the transport of chloride (Cl^-) to the surface and the diffusion of ($CuCl_2^-$) in the solution. An anodic current peak A appeared at a potential of 10 mV and is related to CuCl film form. The electrochemical corrosion parameters of the corrosion potential Ecorr and corrosion current density jcorr obtained by the extrapolation of Tafel lines are listed in Table 1. j_{corr} decreases with increased Schiff base concentration. j_{corr} of the bare copper electrode is 8.13 μA/cm^2 in 0.5 mol/L NaCl solution. *IE* (%) is calculated [20] from the following equation:

$$IE(\%) = \left(\left(j^{\circ}_{corr} - j_{corr}\right) / \, j^{\circ}_{corr}\right) \times 100 \qquad (1)$$

where $j^{\circ}_{\mathrm{corr}}$ and j_{corr} are the corrosion current densities in the absence and presence of Schiff base self-assembled films, respectively.

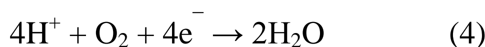
The anodic reactions of copper in the NaCl solution are:



$$Cu + Cl^- \rightarrow CuCl + e^- \qquad (2)$$

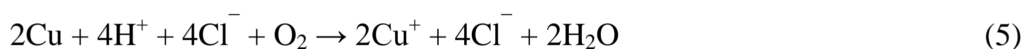
$$CuCl + Cl^- \rightarrow Cu\,Cl_2^- \qquad (3)$$

The cathodic dissolution of copper in NaCl solution is as follows:



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

The total copper corrosion reaction in NaCl solution is:



$$2Cu + 4H^+ + 4Cl^- + O_2 \rightarrow 2Cu^+ + 4Cl^- + 2H_2O \qquad (5)$$

When solid copper dissolves, copper ions chelate with Cl^-. The existence of Cl^- ions subsequently speeds up the dissolution of copper [21-23]. Fig. 3 shows that the anodic polarization is obviously greater than that of the cathode. E_{corr} of the bare copper electrode is -257 mV, and j_{corr} is 8.13 μA/cm^2 (Table 1). When the copper electrode self-assembles in the DPMP-ethanol solution, E_{corr} increases with increasing solution concentration, whereas j_{corr} decreases. As a result, DPMP molecules could inhibit Cl^- from corroding copper. The opportunity for Cl^- to make contact with the copper surface decreases, thereby effectively inhibiting copper dissolution in the anode.

The highest *IE* is about 99.44% in a 0.01 mol/L DPMP-ethanol solution. *IE* increases with increasing self-assembled concentration (Table 1). The DPMP molecules induce changes on the copper electrode surface as well as on the values of j_{corr} and E_{corr}.

3.4. Electrochemical impedance measurements

Fig. 4 shows the Bode plot of the self-assembled film-covered copper electrode (self-assembly time = 2 h) in DPMP-ethanol solutions with different concentrations. From this plot, the equivalent

circuit diagram that fit the impedance spectra can be deduced. The equivalent circuit is shown in Fig. 5. R_s is the solution resistance between the reference and working electrodes. R_t is the charge transfer resistance corresponding to the corrosion reaction at the metal substrate/solution interface. Q_{dl} is the constant phase elements modeling the capacitive loop C_{dl}. W is the Warburg impedance attributed to the mass transport during the corrosion reactions [24-28].

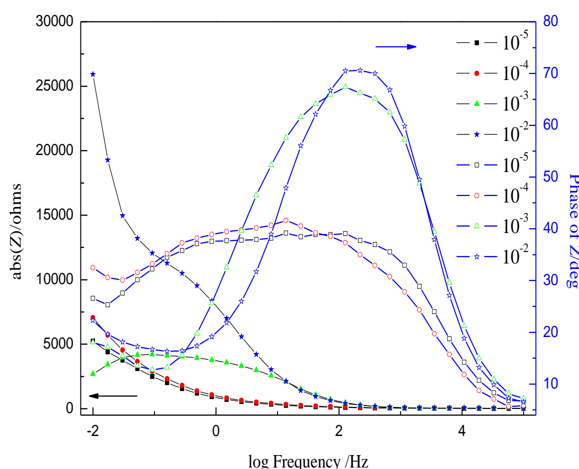


Figure 4. Bode plots of self-assembled films covered copper electrode in different concentrations of DPMP-ethanol solution. Test environment: 0.5 mol/L NaCl solution

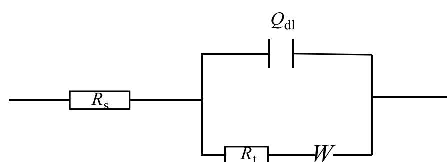


Figure 5. Equivalent circuits to fit the impedance spectra with Warburg (a) and without Warburg (b) impedance

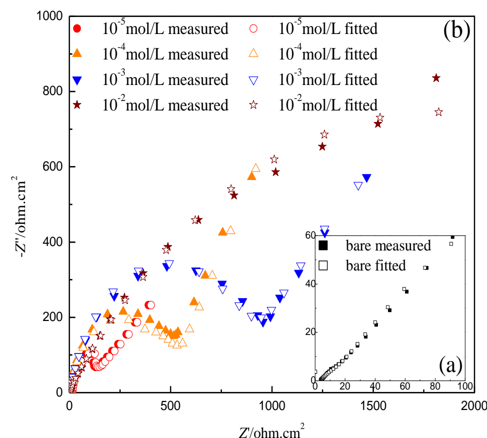


Figure 6. The measured and fitted Nyquist plots of self-assembled films covered copper electrode that self-assembled for 2 h in different concentrations of DPMP-ethanol solution. Test environment, 0.5 mol/L NaCl solution

Fig. 6 is the measured and fitted Nyquist plots of the self-assembled film-covered copper electrode (self-assembly time = 2 h) in DPMP-ethanol solutions with different concentrations. A small capacitive loop in the high-frequency region and a straight line (Warburg) in the low-frequency area are observed in the Nyquist plots of the bare copper electrode. The capacitive loop is attributed to the R_{ct}, whereas the Warburg impedance is attributed to the diffusion of the anodic and cathodic reaction products from the solution to the electrode surface. The equivalent circuit that fits the Nyquist plots is exhibited in Fig. 5.

Table 2. Impedance parameters for copper electrode without and with self-assembled films formed at different DPMP concentrations in 0.5 mol/L NaCl solution

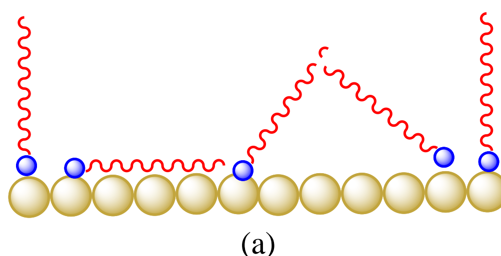
Concentration (mol/L)	R_s (Ω•cm^2)	Q_{dl}		R_t (Ω•cm^2)	W (Ω•cm^2)	IE (%)
		Y_0 (Ω^{-1}•cm^{-2}·s^n)	n(0-1)			
bare	7.09	22.84×10^{-5}	0.7	286.7	3.531×10^{-3}	
10^{-5}	6.90	7.060×10^{-3}	0.8	2772	2.438×10^{-3}	89.66
10^{-4}	6.17	2.372×10^{-5}	0.8	3643	1.198×10^{-3}	92.13
10^{-3}	5.55	1.189×10^{-6}	0.8	6184	7.980×10^{-4}	95.36
10^{-2}	7.31	5.825×10^{-2}	0.8	21360	2.432×10^{-5}	99.87

The values of R_s are similar during the measurements. R_{ct} increases after the electrode surface is covered by the DPMP self-assembled films. The arc radius increases with increasing DPMP concentration, which signifies increased R_t values. The Warburg impedance ceases to exist in a 0.01 mol/L DPMP-ethanol solution. *IE* is calculated from the following equation [29]:

$$IE(\%) = \left(\left(R_t - R_t^{'}\right)/ R_t\right)\times 100 \qquad (6)$$

where R_t and $R_t^{'}$ are the charge transfer resistances in the presence and absence of the DPMP self-assembled films, respectively. The impedance parameters of the copper electrode are shown in Table 2. The corrosion *IE* increases with increasing DPMP-ethanol concentration. The highest *IE* value is 99.87% in a 0.01 mol/L DPMP-ethanol solution. The increased charge transfer impedance is related to the self-assembled films. Therefor, DPMP can inhibit the corrosion of copper in 0.5 mol/L NaCl solution.

3.5. Adsorption studies



(a)

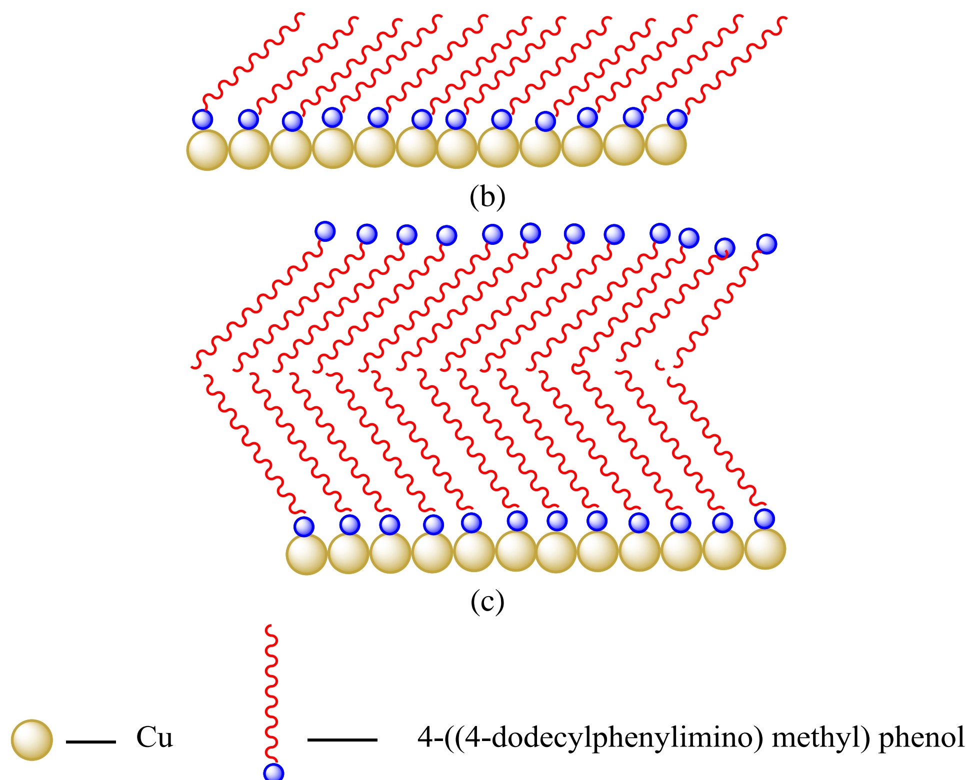


Figure 7. Diagrams of copper in different concentration of Schiff base solution: (a) in a low concentration; (b) in a suitable concentration; (c) in a high concentration

Overall, the results show that self-assembled films of DPMP on a copper surface can be obtained. The hydrophilic groups contain N and O atoms, whose lone pair electrons combine with the empty d orbital of Cu, resulting in stable physical adsorption. At low DPMP concentrations, the molecules are loosely arranged on the copper surface (Fig. 7a). At appropriate concentrations, the self-assembled films are well organized (Fig. 7b). These films may effectively block the oxygen molecules in the solution from making contact with the copper surface, until an efficient anticorrosion effect is attained. However, at excessive Schiff base concentrations, a multilayer structure film is formed (Fig. 7c) [30]. Therefore, the assembly membrane structure is significantly influenced by the Schiff base concentration.

4. CONCLUSIONS

DPMP was a new Schiff base that efficiently inhibited copper corrosion in 0.5 mol/L NaCl solution. Capability of DPMP to inhibit copper corrosion favored the increased adsorption of DPMP molecules on the copper surface. *IE* increased with increasing inhibitor concentration at room temperature. The highest *IE* of DPMP was achieved within a self-assembly time of 2 h in a 0.01 mol/L solution, below which *IE* decreased, as revealed by polarization curve and impedance calculations.

ACKONWLEDGEMENTS
This work was financially supported by the National Natural Science Foundation of China (grant nos. 21276148, 21076119, and 20776081) and the Natural Science Foundation of Shandong Province, China (grant no. ZR2010BM004 and Y2006B37)

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