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Corrosion Inhibition of Cu-Zn Alloys in NaCl Solution Using Isatin

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Abstract

The effect of isatin on the corrosion of Cu-20%Zn-2%Al (alloy I) and Cu-31% Zn (alloy II) in NaCl solution has been studied using different electrochemical techniques [open circuit, potentiodynamic, and impedance spectroscopy (EIS)]. The two copper alloys were used in condenser tubes of heat exchanger for power station plant in Egyptian financial and industrial company Assiut factories. The steady state potential $E_{s.s}$ values increase to more negative values by increasing NaCl concentration which indicated that the destruction of pre-immersion passive film formed on the Cu-Zn surfaces. The corrosion rate of the two alloys decreased by increasing concentration of isatin, which indicated the effect of isatin as a corrosion inhibitor. The inhibition efficiency of isatin in 0.1M NaCl solution was 88.5 % in alloy I and 73.9 % in alloy II at a concentration 0.01M of isatin. The significant inhibition processes due to the adsorption characteristics of isatin on the two alloys surface. The chemical composition of alloy I was more resist to corrosion in NaCl solution than the chemical composition alloy II due to presence of Al.

Keywords: Cu-Zn alloys, corrosion, isatin, inhibition, NaCl.

1. Introduction

The excellent electrical and thermal conductivity combined with a good mechanical workability of copper and its alloys enable it to use in industrial applications. The chemical industries employ copper and its alloys extensively for condensers evaporator's heat exchangers fractionating columns etc. The corrosion resistance of copper and its alloys is often in sufficient when a contact with aggressive solutions. Indeed, in some environments containing ions such as chloride sulfate or nitrate passive film breakdown occur leading the propagation of corrosion [1-5]. Type's Cu-31%Zn and Cu-20%Zn-2%Al alloys are used in power station condensers tubes in Egyptian Financial and Industrial Company (EFIC) Assiut factories Egypt in main and auxiliary condensers. In these condensers there is steam water in shell side and cooling water in tube side (River Nile water) most of corrosion occurs in tube side due to the contamination of cooling water in the field of industrial. Deposits and films which accumulate and grow on the tubes inside surface effect on heat transfer capacity and in turn its ability to condense steam. The normal medium in circulation cooling is neutral medium which is predominant in most cases and all time of operation therefore aim of the work is studding the corrosion and inhibition of two copper alloys in neutral medium for more protection in these tubes of condenser and long period of operation without re tubing of condenser then low cost. Some of inorganic inhibitors are used to inhibit copper and its alloys [6-7]. Many of organic inhibitors are used azoles amines amino acids [8] furthermore a great deal of interest in the use of natural product as a corrosion inhibitor, [9-15]. The primary step in the action of organic corrosion inhibitors is usually adsorption at metal solution interface. Adsorption process depends upon the electronic molecules, the chemical composion of the solution, and nature of the metal surface, temperature of the reaction and on the electrochemical potential at the metal solution interface [16]. Adsorption can be physisorption or

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chemisorption or both [17]. Physisorption is due to electrostatic attractive forces between inhibiting organic ions or dipoles and the electrical charged surface metal but chemisorption is due to the interaction between uncharged electron pairs of ñ electrons with the metal in order to form a coordinate type of bond. It takes place in presence hetero atoms (P, Se, S, N, O) with the lone pair of electrons and /or aromatic ring in the adsorbed molecules [18-21]. Isatin which has two carbonyl groups in the 2 and 3 position acts as an indole derivative. The carbonyl group in 3 positions is very reactive behaves like acetone that; the other has amidic behavior [19]. The infrared evidence supports the structure for isatin and gives no evidence for the enol form [22].

Isatin not only exhibit wide range of biological activities such as anticonvulsant, analgesic, anti-inflammatory, antidepressant, pro-apoptotic, cytotoxicity, antioxidant, and antimicrobial but also act as efficient corrosion inhibitors [23–27]. Isatin has been successfully used as inhibitors for the present system in acidic medium in the same laboratory [28]. Also, a little attention has been devoted for the development of isatin as corrosion inhibitor in NaCl. Therefore, the aim of this paper is to further explore the use of isatin as corrosion inhibitor for Cu-31%Zn and Cu-20%Zn-2%Al surfaces in NaCl solution by open circuit potential, potentiodynamic polarization, and electrochemical impedance spectroscopy measurements. Additionally, the effect of Al on the resistance of alloy to corrosion, the role of presence isatin improving the passive film resistance to corrosion in presence of Cl anion, and the stability of passive film under different conditions by different techniques.

2. Experimental

Experiments were carried out in NaCl solution in absence and presence of different concentrations of isatin. All solutions were prepared by using double distilled water. The chemicals (isatin and sodium chloride) are A. R grade. A new polished electrode is used for each run. All experiments were carried out at room temperature (25□C) and under purified nitrogen gas. The two alloys I and II were used in this study as working electrodes. The chemical composition by (wt %) of alloy (I) is 78% Cu, 20% Zn and 2% Al and the chemical composition by (wt %) of alloy (II) is 69% Cu and 31% Zn. These alloys were prepared by Central Metallurgical R& Institute Cairo Egypt. The ingots were machined in the form

of short rods, each 20 mm in length and 5 mm in diameter. Each working electrode was constructed and treated following the procedure described previously [29]. A Pt sheet was used as a counter electrode. The potential measurements were measured against Ag/AgCl electrode as a reference electrode. The polarization measurements were performed by using computerized potentiostate (Radiometer model Volta Lab 40) and Volta Master 4 software in the range of -1000 to 2000 mV with scan rate 1 mV/sec. The impedance measurements were performed by using the same instrument between the range 10−3 Hz to 100 KHz, at amplitude of 5mV

3.Results and discutios

3.1. Effect of NaCl Concentration

The steady state values of the two Cu-Zn alloys immersed in different concentrations of NaCl solutions were determined as a function of time by using the open circuit potentials technique. The observation of these values that there is approached to more negative values by increasing NaCl concentration which indicate that the destruction of pre-immersion passive film formed on the Cu-Zn surfaces. The data in Fig.1 shows the straight line relationship between Es.s and NaCl in different concentration according the following equation:

$$Es.s = a - b \ Log \ [NaCl] \qquad (1)$$

Where the symbols a and b are constants and depending upon the chemical composition of the Cu-Zn alloy. The results show there is a negative shift in Es.s values and therefore there is anodic (metal dissolution) reaction then NaCl accelerate the anodic dissolution. This could be attributed to direct participation of adsorbed Cl- ions on ionization of the alloy. In these results one can conclude that, the corrosion of Cu-Zn alloys in NaCl solution is determined by polarization of the anodic controlling reaction, i.e., the corrosion process is anodically controlled. The data in Fig. 1 shows also that the steady state potentials of alloy II are more negative than those of alloy I. These results indicate that alloy I suffers les corrosion than alloy II and this observation may be attributed to the presence of Al in alloy I. Aluminium is known as promoter of β phase in Cu-Zn alloys and considered at least 6 times effective than Zn. In other words, 1% Al addition is as effective as 6% of Zn addition. Addition of 0.35%

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Al to Cu36%Zn the macro structure shows a marginal decrease ingrain size while the microstructure is completely transformed. The grain is equated and has a matrix containing β with α needle dispersed with in the matrix. This is a typical structure of high strength yellow brass which contains much more aluminium [30]
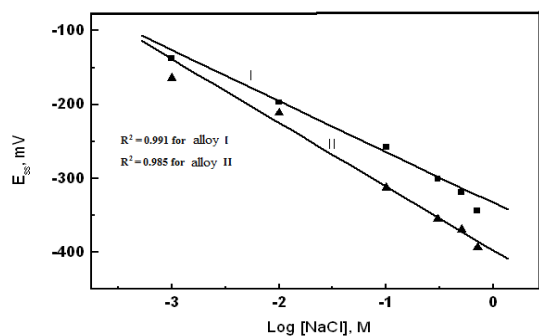

Fig. 1 Variation of steady state potential (Es.s) for alloys (I, II) with the) with the log concentration of NaCl.

Fig 2 indicates the potentiodynamic polarization curve for the two alloys (I, II) between the range - 1000 to 2000 mV in different concentrations of NaCl solution in this finger an increase in NaCl concentration there is also increases in the corrosion rate for the two alloys.
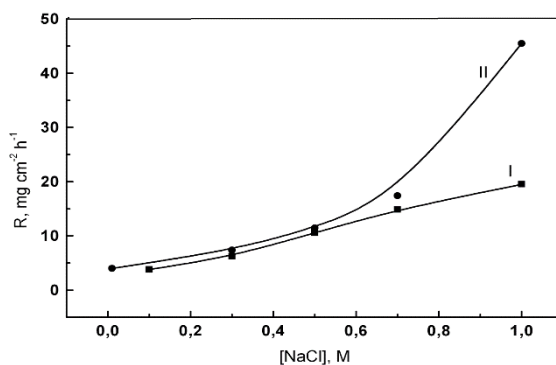

Fig. 2   Corrosion rates as a function of NaCl concentrations for alloys (I, II).

The adsorbed Cl- ions on the alloys surface cause the ionization of the metal atoms and the order of the corrosion resistance of these alloys is: I >II, this sequence can be argued to the presence of Al in alloy (I). Pourbaix diagram for Al [31], give a significant rule for these results and a stable Al2O3 passive film formed in the range of (pH = 4 − 8) and in the potential range (-1.8 to 1.1 V)

3.2. Effect of isatin Concentration

Effect of isatin on the anodic and cathodic polarization of Cu-Zn alloys (I, II) in 0.1 M NaCl solution is investigated as shown in Figs. 3a and 3b. The anodic and cathodic polarization of the Cu-Zn electrode (for two alloys) in 0.1 M NaCl solution containing different concentrations of isatin is shifted to less negative values from - 0.360 V to - 0.325 V in alloy I and from - 0.400 V to - 0.250 V in alloy II, with decreasing current density but it is markedly in cathodic polarization specially in alloy (II) Fig 3.b.
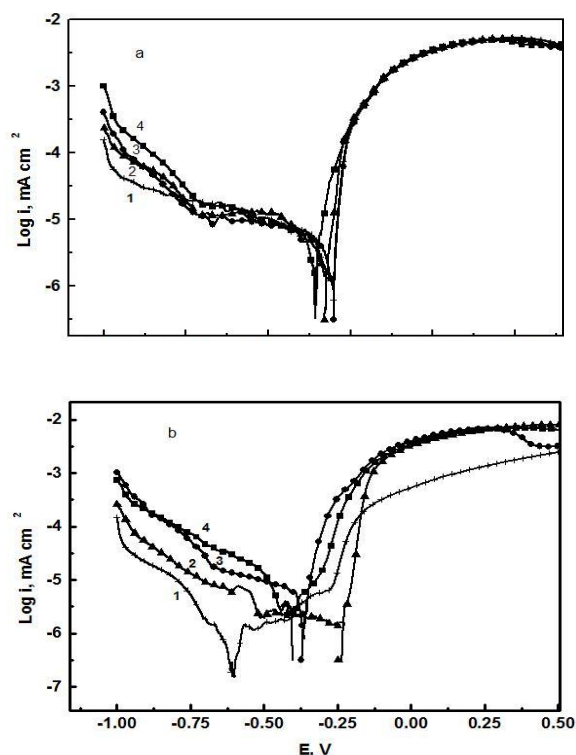

Fig. 3 Polarization curves for (a) alloy I and (b) alloy II in 0.1 M NaCl solution in presence of different concentrations of isatin: 1) 0.0, 2) 0.003, 3) 0.005, 4) 0.01 M.

This indicates that the presence of isatin as inhibitor in the corrosive medium increases the anodic and cathodic overpotential and also decreases the corrosion current (icorr). These changes increase with increase isatin concentration. This behavior supports the inhibition function of isatin for both cathodic and anodic polarization. Figs. 3a and 3b also indicate the difference between alloy (I) and alloy, (II) to resist corrosion due to Al in alloy (I) as the same argued before.

The data in Fig. 4 shows the relation between the corrosion rate and the concentration of isatine in 0.1M NaCl solution for the two alloys I and II this relation indicate that there is decreasing of corrosion rate by increasing of isatin concentration in both alloys. The two upper straight horizontal lines a-b and c-din this figure is the corrosion rates of the two alloys I and II respectively in 0.1M and absence of isatin. This result is suggesting that isatin is acting as corrosion inhibitors for the two Cu-Zn alloys in NaCl solution.
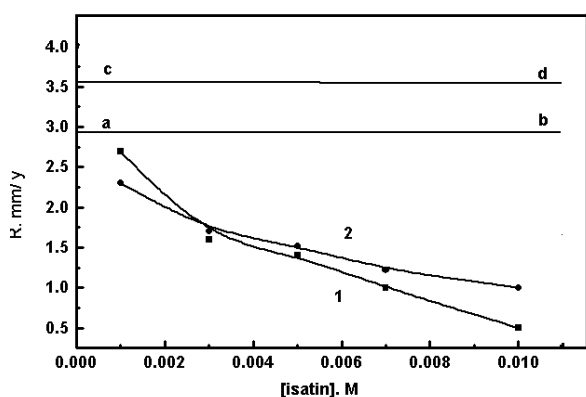

Fig. 4 Corrosion rate for two alloys in 0.1 M NaCl as a function of isatin concentrations: 1) alloy I, and 2) alloy II, a-b and c-d lines, are the values of blank corrosion rate of two alloys I and II respectively in 0.1 M NaCl

The data in Fig. 5 illustrate the inhibition efficiency (IE) which can be calculated from polarization measurements according to the following equation:

$$IE (\%) = [1 - (R1/R2)] \times 100 \qquad (2)$$

The R1 and R2 are the corrosion rates in the presence and absence of isatin respectively. From the data in this figure there is a directly proportional between the inhibition efficiency of isatin and its concentration, which is a straight-line relationship between IE% and isatin at different concentration. These results can be explained due to the competitive adsorption between isatin molecules and the aggressive Cl- anion on the electrode surface which retards their corresponding destructive action [32]
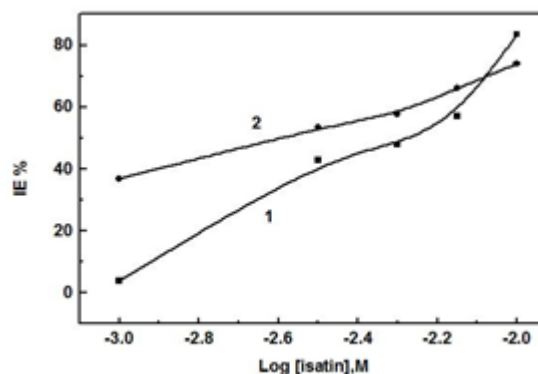

Fig. 5 Inhibition efficiency for two alloys in 0.1 M of NaCl as a function of isatin concentrations: 1) alloy I and 2) alloy II.

The inhibitive molecule of isatin may be incorporated into the passive layer on the alloy surface, forming an improved barrier against the aggressive ions. Passive layer of isatin which content in the surface of alloys increase by increasing isatin concentration which leads to increase the corrosion resistance [33, 34]. The inhibition efficiency of isatin in alloy (II) is more than alloy (I) which contain extend self-passivation due to presence of Al.

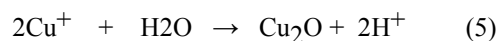3.4. Mechanism of Corrosion Inhibition

The film formation and kinetics mechanism of anodic dissolution of brass in neutral chloride solution can be expressed by the following reactions [35]. In the initial corrosion stage, zinc forms ZnO as a result of
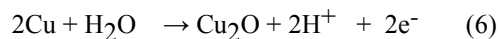
$$Zn^{+2} + H_2O \rightarrow ZnO + 2H^+ \qquad (3)$$

or

$$Zn + H_2O \rightarrow ZnO + 2H^+ + 2e^- \qquad (4)$$
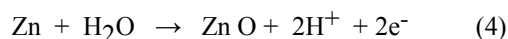
Then copper forms Cu2O as a result of

$$2Cu^+ + H2O \rightarrow Cu_2O + 2H^+ \qquad (5)$$

or

$$2Cu + H_2O \rightarrow Cu_2O + 2H^+ + 2e^- \qquad (6)$$

After the surface has become covered by both ZnO and Cu2O, the CuCl is formed on the surface by the reaction

$$Cu^+ + Cl^- \rightarrow CuCl \qquad (7)$$

Which can then undergo the disproportionation reaction [36]

$$2CuCl \rightarrow Cu + CuCl_2 \qquad (8)$$
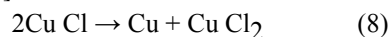
or dissolve with the formation of CuCl2 complexes [37,38] via

$$Cu\,Cl + Cl^- \rightarrow Cu\,Cl_2^- \qquad (9)$$
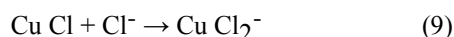
The inhibitive action of isatin (hetero organic compound) which containing N and O atoms leads to the formation of a co-ordination bond between the metal and the lone pair of electrons present in the hetero atoms then the extent of inhibition can be enhanced [39]. These factors play the vital role in the adsorption of the inhibitor on the metal surface.

3.4. Electrochemical Impedance Investigations of Isatin Inhibition

The impedance complex diagrams (Nyquist plot) for two alloys I and II in 0.1 M NaCl solution investigated in Figs.6a and 7a at the corrosion potential (Ecorr). There is a semicircle type appearance in these diagrams at high frequency followed by a straight line at low frequency. The semicircles at high frequency indicate that the corrosion of two alloys is mainly controlled by charge transfer process but straight lines at lower frequency indicate a limited process of mass transfer correspond to the ions diffusion in the structure (Warburg impedance) as shown in previse equations mechanism of corrosion.
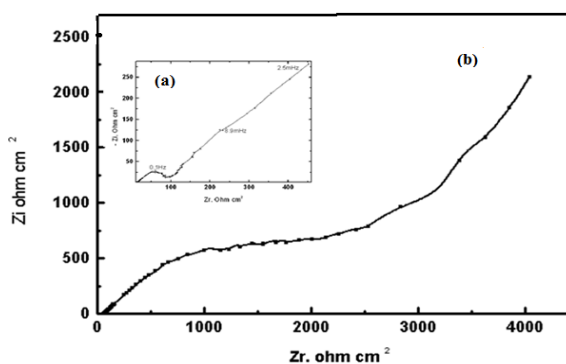

Fig. 6 Nyquist plot for alloy I in (a) 0.1 M NaCl and (b) 0.1 M NaCl and 0.007 M of isatin

The impedance complex diagram of two alloys (I, II) in 0.1 M of NaCl in presence of isatin was shown in Figs. 6b and 7b. These impedance diagrams show that in the presence of isatin there is a decrease in the charge transfer process for two alloys and the semicircle disappear at higher frequency.

The high resistance magnitudes in Fig 6b and 7b are due to the presence of a compact and protective film of inhibitor molecules. The high resistance in Fg

7b than in fig 7a due to the high inhibition efficiency of alloy (II) thane alloy (I). Electrochemical theory shows that the reciprocal of the charge transfer resistance is proportional to the corrosion rate [40].
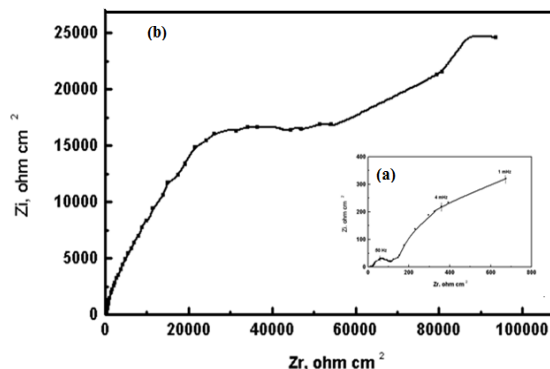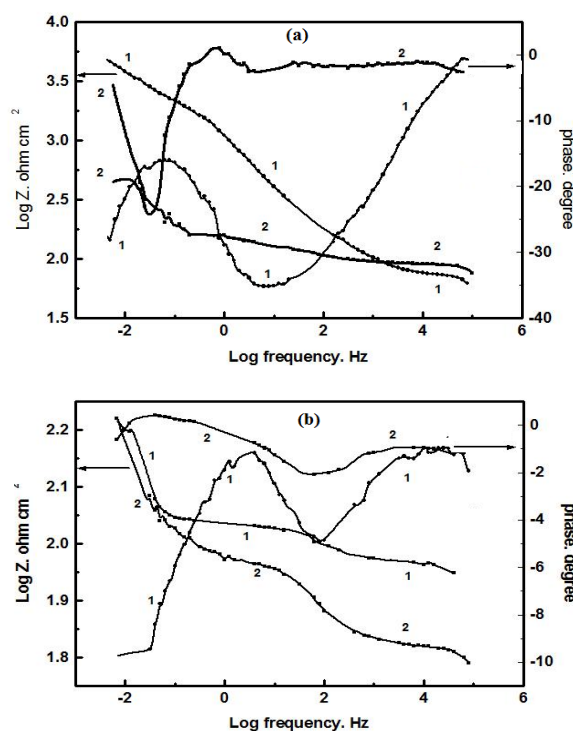

Fig. 7 Nyquist plot for alloy II in (a) 0.1 M NaCl and (b) 0.1 M NaCl and 0.007 M of isatin.

The Bode plots for the relation between log ω versus θ in Figs.8a, and fig.8b for the two alloys I and

Figs. 8 Bode plot (a) for alloy I (I) 0.1 M NaCl and (2) 0.1 M NaCl + 0.01 M isatin (b) for alloy (II) 0.1 M NaCl and (2) 0.1 M NaCl + 0.01 M isatin

II in 0.1 M NaCl solution in presence and absence of isatine indicate there is a one relaxation process showed by one phase maximum.These results also indicate that there is one charge transfer processes

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taking place at the alloy-electrolyte interface due to the diffusion of different component (Cl- and isatin) at the alloy electrolyte interface.

The impedance values in the relation Bode plot (log ω versus |Z|) show also that the presence of inhibitor molecules, which may be due to the adsorption of different corrosion products and inhibitor species (isatin) on the surface of Cu-Zn alloy the slope of the curve in presence of isatin is upper thane unity at lower frequency which indicate that the resistive behavior [41-42]. Fig.9a and Fig.9b indicate the comparison of impedance in Bode plot for two alloys I and II in 0.1 M NaCl solution in presence of 0.005 M isatin which is increase in the order: II >I and these results and this sequence due to the high inhibition efficiency of alloy II than alloy I in presence of isatine as discussed above.
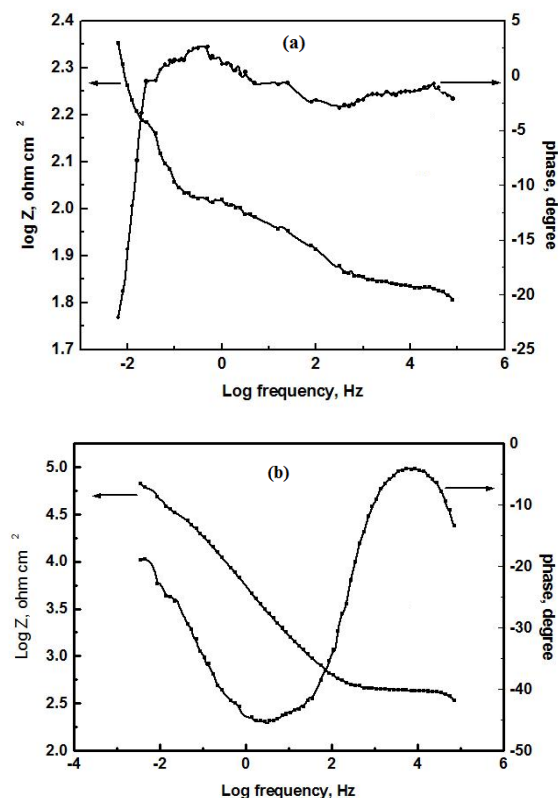

Fig. 9 Bode plot for(a) alloy I and (b) alloy II in 0.1 M NaCl + 0.007M isatin.

Figure (10a, b) shows the important relation in electrochemical impedance between the capacitance of the alloy solution interface and the time in 0.1 M NaCl solution in the absence and presence of isatin. It appears that the capacitance of alloy-solution

interface in presence of isatin is lower than that in 0.1 M NaCl solution without isatin. This behavior is mainly due to the adsorption of the isatin molecules at the alloy solution interface.
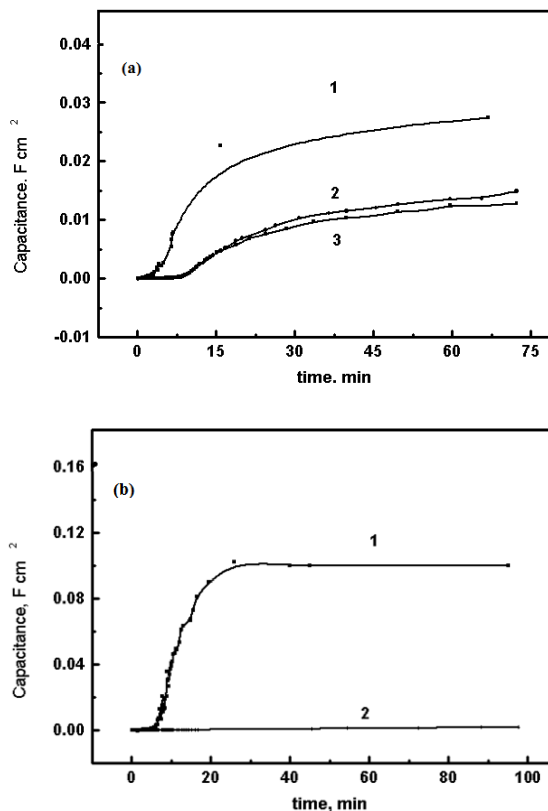

Fig. 10 Dependence of the capacitance on the time for (a) alloy I and (b) alloy II in: 1) 0.1 M NaCl, 2) 0.1 M NaCl + 0.007 M isatin, 3) 0.1 M NaCl + 0.01 M isatin,

4.conclusion

Isatin acts as a good corrosion inhibitor for the two copper alloys (I, II) in 0.1 M NaCl for both cathodic and anodic polarization. The corrosion rate of alloy I in NaCl solution and absence of isatin is lower than in alloy II due to presence of Al. The inhibition efficiency values increase with the inhibitor concentrations and the maximum inhibition efficiency is about 88.5 % in alloy I and 73.9 % in alloy II. The negative shift in the values of corrosion potential (ECorr) indicated that the presence of isatin as inhibitor in the corrosive medium increases the anodic and cathodic overpotential and also decreases the corrosion current (icorr). EIS measurements show that the charge resistance increases and double layer

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capacitance decreases in the presence of isatin inhibitor indicating the adsorption of the inhibitor at surfaces of Cu-Zn alloys.

5- conflict of interests

The author declare that have no conflicts of interested

6- Acknowledgment

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