

Corrosion inhibition of Al-Mg alloy by gentisic acid

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Gentisic acid was studied for possible use as an Al-2.5Mg alloy corrosion inhibitor in a 0.5 mol dm⁻³ NaCl solution. Investigations were performed on a rotating disc electrode in a quiescent solution at different electrode rotation rates and electrolyte temperatures. The results indicate that, the addition of gentisic acid inhibits corrosion of Al-2.5Mg alloy, and the inhibition efficiency increases with increasing inhibitor concentration but decreases with increasing electrode rotation rate and electrolyte temperature. The investigated compound acts as a cathodic-type inhibitor and the inhibition is ascribed to the adsorption of the inhibitor onto the electrode surface. The adsorptive behaviour of gentisic acid follows the Freundlich adsorption isotherm.

Keywords: Al-Mg alloy, Corrosion inhibition, Gentisic acid, Rotating disc electrode, Adsorption, Activation energy

The application of aluminium and its alloys in different areas has been constantly increasing. Therefore, Al-Mg alloys have found wide application in shipbuilding, chemical and food industry. Good corrosion resistance of Al and its alloys is attributed to the presence of a thin protective surface oxide film. This oxide film is relatively stable in the range from pH 4 to 9, but in the presence of aggressive anions, even in this pH range, the protective layer can be locally destroyed and the corrosive attack can take place. One of the methods of metal and alloy corrosion protection is the application of corrosion inhibitors, but many commercial inhibitors have toxic properties and have adverse effects on the environment; so intensive research is in progress to find new ecologically acceptable inhibitors¹⁻⁶. This study is a continuation of research of corrosion inhibition of Al-2.5 Mg alloy with compounds that exist in Mediterranean plant *Rosmarinus officinalis* L. The aqueous extract of *Rosmarinus officinalis* L. contained neutral and acid phenolic compounds^{1,7,8}. Investigations of corrosion inhibition of Al-2.5Mg alloy with a neutral fraction of aqueous extract have shown that the dominant organic compound was (+)-catechine, and that this compound showed good cathodic corrosion inhibition for this alloy^{1,9}. Acidic subfractions of aqueous extract also have shown corrosion inhibition characteristics^{7,8}. Analyses of acidic extract have shown the presence of different phenolic acids. One of them was sinapinic acid and

the investigations with the pure sinapinic acid have shown that this acid acts as cathodic corrosion inhibitor for Al-2.5Mg alloy¹⁰.

The aim of present work is to study the effect of gentisic acid on the corrosion of Al-2.5Mg alloy in a 0.5 mol dm⁻³ NaCl solution using polarization and impedance techniques. Gentisic acid belongs to the group of phenolic acids, substances that naturally occur in fruit, vegetables, nuts, seeds, flowers and some herb beverages^{11,12}, and it is also found in acidic fraction of acidic aqueous extract of *Rosmarinus officinalis* L. This phenolic acid shows good antioxidant and also antibacterial activity^{12,13}.

Experimental Procedure

A disc electrode made out of the Al-2.5Mg alloy suitable for a rotating disc system was prepared by placing a cylinder alloy sample in a Teflon holder, leaving 0.5 cm² area exposed. Table 1 shows the chemical composition of the alloy.

Before each experiment, the electrode surface was abraded with different grades of emery papers, polished with alumina slurries (0.5 and 0.05 μm), degreased in ethanol, rinsed with doubly distilled water and left a few minutes in air to form the natural oxide film before being immersed in the solution. Electrochemical measurements were carried out in a conventional three electrode cell equipped with double-jacketed device allowing thermostatic experiments to be performed. A saturated calomel

Table 1—Chemical composition of the Al-2.5Mg alloy

| Element | Composition (wt. %) |
|---------|---------------------|
| Si | 0.19 |
| Fe | 0.40 |
| Cu | 0.06 |
| Mn | 0.27 |
| Mg | 2.59 |
| Cr | 0.02 |
| Zn | 0.04 |
| Ti | 0.02 |
| rest | 0.1 |
| Al | balance |

electrode (SCE) with a Luggin capillary was used as a reference and Pt plate as a counter electrode. All solutions were prepared from pro-analysis grade reagents. The basic solution was 0.5 mol dm^{-3} NaCl solution to which gentisic acid was added in concentrations of 1×10^{-5} , 5×10^{-5} , 1×10^{-4} , 5×10^{-4} and $1 \times 10^{-3} \text{ mol dm}^{-3}$. The pH values of all investigated solutions were adjusted to 6 using sodium hydroxide solution.

Figure 1 shows the structural formula of gentisic acid. The experiments were performed in a quiescent solution in the temperature range from 20 to 50°C , and at the electrode rotation rates of 150 and 2000 rpm.

The potentiodynamic polarization measurements were performed using a potentiostat (PAR M 273A), with the scanning rate of 0.2 mV s^{-1} .

The polarization resistance, R_p , was determined from the slope of polarization curves obtained by measurements in the potential range from -10 to $+10 \text{ mV}$ near the corrosion potential at a scanning rate of 0.2 mV s^{-1} . Electrochemical impedance spectroscopy (EIS) was carried out with a lock-in amplifier (PAR M5210), at the open circuit potential (E_{OCP}) in a quiescent solution. The a.c. amplitude was $\pm 10 \text{ mV}$ and the frequency range studied was from 50 kHz to 30 mHz. The impedance diagram is given in the Nyquist representation.

Results and Discussion

Figure 2 shows the potentiodynamic polarization curves for Al-2.5Mg alloy in a 0.5 mol dm^{-3} NaCl solution without and with the addition of gentisic acid in different concentrations at 20°C . The addition of gentisic acid induces changes in the polarization behaviour of the alloy examined. Changes observed in the polarization curves after addition of the inhibitors are usually used as criteria to classify inhibitors as cathodic, anodic or mixed¹⁴. In the present case, the modifications caused by addition of gentisic acid to the NaCl solution are a negative shift on the corrosion

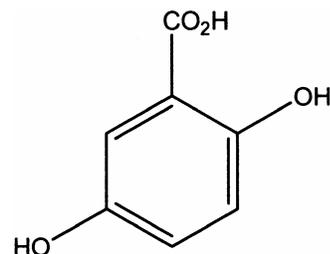


Fig. 1—Structural formula of gentisic acid (2,5-dihydroxybenzoic acid).

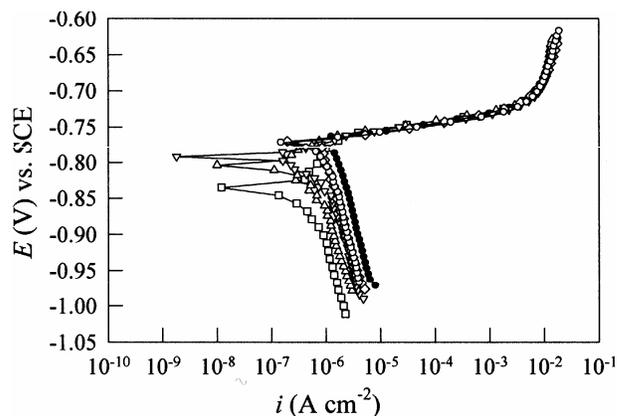


Fig. 2—Potentiodynamic polarization curves for the Al-2.5Mg alloy in quiescent 0.5 mol dm^{-3} NaCl solution at $T = 20^\circ\text{C}$, without (\bullet) and in the presence of gentisic acid in the concentration of $1 \times 10^{-3} \text{ mol dm}^{-3}$ (\square), $5 \times 10^{-4} \text{ mol dm}^{-3}$ (Δ), $1 \times 10^{-4} \text{ mol dm}^{-3}$ (∇), $5 \times 10^{-5} \text{ mol dm}^{-3}$ (\diamond) and $1 \times 10^{-5} \text{ mol dm}^{-3}$ (\circ).

potential (E_{corr}) and a leftward displacement in the cathodic branch of the curves. In the anodic domain the i - E characteristics remained almost the same. These changes indicate that the gentisic acid acts as a cathodic corrosion inhibitor of Al-2.5Mg alloy under the given conditions. Lowering the inhibitor concentration decreases the changes mentioned. The effect of the electrode rotation rate on the polarization curves is presented in Fig. 3. Increase in electrode rotation rate leads to some increase in cathodic current density and to reduction of negative shift of corrosion potential caused by addition of inhibitor, which shows the lowering of the corrosion inhibition effect.

The surface coverage (Θ) and inhibition efficiency (η) were calculated using Eqs (1) and (2):

$$\Theta = \frac{i_{\text{corr}} - (i_{\text{corr}})_{\text{inh.}}}{i_{\text{corr}}} \quad \dots (1)$$

$$\eta = \Theta \times 100 \quad \dots (2)$$

where i_{corr} and $(i_{\text{corr}})_{\text{inh}}$ represent corrosion current densities in the absence and presence of inhibitor.

Table 2 shows the values of corrosion current density, surface coverage and inhibition efficiency that were determined from the corrosion current density, at different electrode rotation rates and temperature of 20°C. From Table 2 it is evident that the addition of the gentisic acid decreases the corrosion current, and this decrease is more pronounced with the increase of inhibitor concentration. So, the efficiency of gentisic acid as corrosion inhibitor for the Al-2.5Mg alloy increases with the increase in its concentration but decreases with the increase in electrode rotation rate.

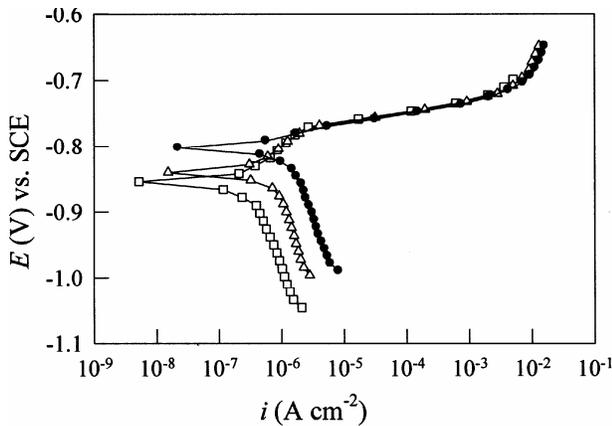


Fig. 3—Potentiodynamic polarization curves for the Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl solution in the presence of gentisic acid in the concentration of 1 × 10⁻³ mol dm⁻³, at 20°C and different electrode rotation rates: ω = 0 rpm (□), ω = 150 rpm (Δ) and ω = 2000 rpm (●).

Table 2—Corrosion current density, surface coverage and inhibition efficiency for the Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl solution with addition of different concentration of gentisic acid

| C (inhibitor) (mol dm ⁻³) | ω (rpm) | i _{corr} (μA cm ⁻²) | Θ | η (%) |
|--|------------|---|--------|----------|
| 0 | 0 | 1.44 | - | - |
| 1×10 ⁻⁵ | | 0.87 | 0.3958 | 39.58 |
| 5×10 ⁻⁵ | | 0.77 | 0.4653 | 46.53 |
| 1×10 ⁻⁴ | | 0.67 | 0.5347 | 53.47 |
| 5×10 ⁻⁴ | | 0.56 | 0.6111 | 61.11 |
| 1×10 ⁻³ | | 0.46 | 0.6806 | 68.06 |
| 0 | 150 | 2.28 | - | - |
| 1×10 ⁻⁵ | | 1.55 | 0.3202 | 32.02 |
| 5×10 ⁻⁵ | | 1.41 | 0.3816 | 38.16 |
| 1×10 ⁻⁴ | | 1.27 | 0.4430 | 44.30 |
| 5×10 ⁻⁴ | | 1.08 | 0.5263 | 52.63 |
| 1×10 ⁻³ | | 0.93 | 0.5921 | 59.21 |
| 0 | 2000 | 2.84 | - | - |
| 1×10 ⁻⁵ | | 2.28 | 0.1972 | 19.72 |
| 5×10 ⁻⁵ | | 2.13 | 0.2500 | 25.00 |
| 1×10 ⁻⁴ | | 1.98 | 0.3028 | 30.28 |
| 5×10 ⁻⁴ | | 1.79 | 0.3697 | 36.97 |
| 1×10 ⁻³ | | 1.58 | 0.4437 | 44.37 |

Since the inhibition works through the mechanism of adsorption of the inhibitor molecules on the metal surface, the use of adsorption isotherms provides useful insight into the inhibition mechanism. In order to study the relationship between inhibitor adsorption on the electrode surface and its concentration, the degree of surface coverage Θ, defined by Eq. (1), was plotted as a function of the gentisic acid concentration (Fig. 4). The Freundlich adsorption isotherm¹⁵ is:

$$\Theta = KC^n \quad \dots (3)$$

where 0 < n < 1, or

$$\ln \Theta = \ln K + n \ln C \quad \dots (4)$$

K and C represent the equilibrium constant of adsorption and the inhibitor concentration. A plot of ln Θ against ln C gives a straight line of intercept ln K. Linear correlations were obtained, suggesting that the Freundlich adsorption isotherm was obeyed consistently.

The equilibrium constant of adsorption, K, is related to the standard free energy of adsorption, ΔG_{ads}^o by Eq. (5):

$$K = \frac{1}{55.5} \exp\left(\frac{-\Delta G_{ads}^o}{RT}\right) \quad \dots (5)$$

where R is the universal gas constant, the value of 55.5 is the concentration of water in the solution, and T is the absolute temperature. The value of standard free energy of adsorption indicates the adsorption mechanism of organic compound on the metal surface. The calculated value of ΔG_{ads}^o was -10.793 kJ mol⁻¹, which indicates physical adsorption of organic compound on the metal surface.

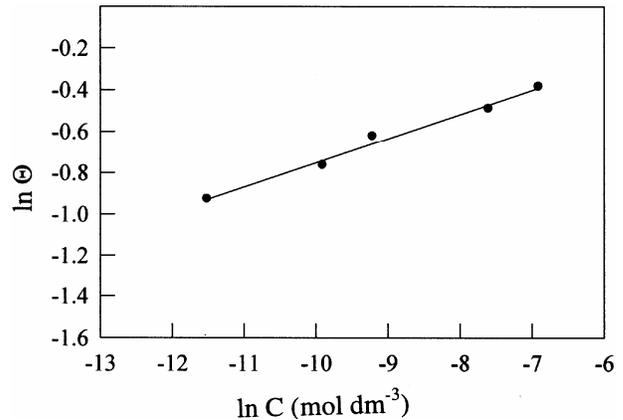


Fig. 4—Adsorption isotherm for gentisic acid.

The effect of temperature on corrosion rate of Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl without and in the presence of gentisic acid is studied in the temperature range 20-50°C. The increase in electrolyte temperature leads to increase in corrosion currents for the measurements in uninhibited and inhibited NaCl solution. Inhibition efficiency depends upon the temperature and decreases with temperature.

The dependence of corrosion rate on temperature can be regarded as Arrhenius-type process, which can be described by the Eq. (6):

$$i_{\text{corr}} = A \exp\left(-\frac{E_a}{RT}\right) \quad \dots (6)$$

where i_{corr} is the corrosion current density, A is the Arrhenius preexponential constant and E_a is the apparent activation energy.

Arrhenius plots (i.e., the logarithms of the corrosion current density against $1/T$) are presented in Fig. 5. Straight lines are obtained for noninhibited and inhibited NaCl solution. The values of E_a were determined from the slopes of these lines and calculated to be: $E_{a1} = 24.94 \text{ kJ mol}^{-1}$ in a noninhibited NaCl solution and $E_{a2} = 42.00 \text{ kJ mol}^{-1}$ in the presence of the highest concentration of gentisic acid.

Generally, the inhibitive additive cause a rise in activation energy value when compared to the base solution and could be interpreted by physical adsorption^{16,17}. Also physical adsorption is related to lower values of the activation energy (30-50 kJ mol⁻¹)¹⁶, which is agreeable to the presented results.

Figure 6 shows linear parts of polarization curves obtained by the linear polarization method for Al-2.5Mg alloy in a 0.5 mol dm⁻³ NaCl solution in the presence of gentisic acid in concentration $1 \times 10^{-3} \text{ mol dm}^{-3}$ at different electrode rotation rates. The increase in electrode rotation rate leads to a reduced slope of linear parts of the curve i.e. reduced values of the polarization resistance. The increase of the electrolyte temperature has similar negative effect on the values of the polarization resistance.

The values of the polarization resistance, R_p , were determined from the slope of the linear polarization curves near E_{corr} :

$$R_p = \left(\frac{dE}{dI}\right)_{E=E_{\text{corr}}} \quad \dots (7)$$

R_p values were used to calculate the inhibiting efficiency, η , using the equation:

$$\eta = \left(\frac{R_{pi} - R_p}{R_{pi}}\right) \times 100 \quad \dots (8)$$

where R_p and R_{pi} are the polarization resistances without and with the addition of inhibitor.

The values of the polarization resistance and the calculated inhibitor efficiency, and surface coverage, are shown in Table 3. Generally, R_p values increase with increasing inhibitor concentration but decrease with increasing electrode rotation rate. Also, increasing temperature leads to decreasing R_p values. The values of inhibiting efficiency, calculated from the values of polarization resistances, fit well

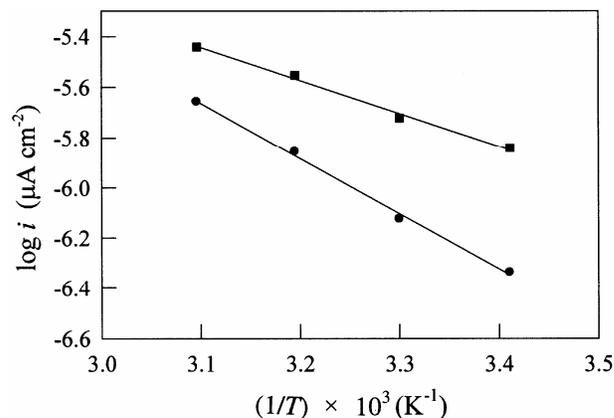


Fig. 5—Activation energy determination for the Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl solution without (■) and in the presence of gentisic acid in the concentration of $1 \times 10^{-3} \text{ mol dm}^{-3}$ (●).

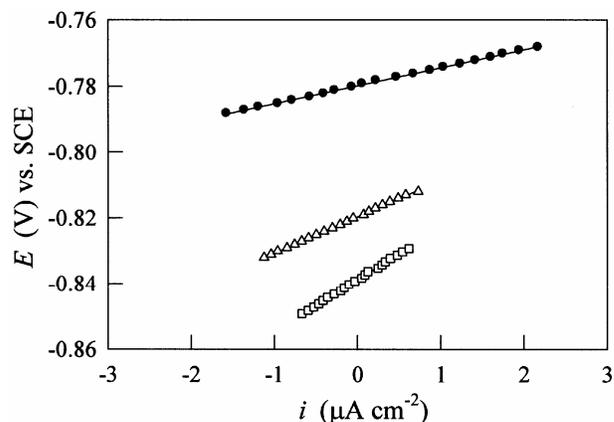


Fig. 6—Linear polarization curves for determination of polarization resistance of the Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl solution in the presence of gentisic acid in the concentration of $1 \times 10^{-3} \text{ mol dm}^{-3}$ at 20°C and at the electrode rotation rates $\omega_1 = 0 \text{ rpm}$ (□), $\omega_2 = 150 \text{ rpm}$ (Δ) and $\omega_3 = 2000 \text{ rpm}$ (●).

with the values calculated from the corrosion current densities obtained from potentiodynamic polarization measurements.

The electrochemical impedance spectroscopy measurements were carried out in a quiescent NaCl solution without and with the addition of different concentrations of gentisic acid at 20°C. The results are shown in Fig. 7. As can be seen, two loops were observed in the Nyquist diagram for the measurements in a NaCl solution in the absence and in the presence of gentisic acid in concentration 1×10^{-5} mol dm⁻³ and 5×10^{-5} mol dm⁻³: the capacitive loop in the high-frequency range and an inductive loop in the low-frequency domain. The time constant

Table 3—Polarization resistance, surface coverage and inhibition efficiency for the Al-2.5Mg alloy in a 0.5 mol dm⁻³ NaCl solution without and with addition of different concentration of gentisic acid at 20°C and different electrode rotation rates

| C (inhibitor) (mol dm ⁻³) | ω (rpm) | R_p (k Ω cm ²) | Θ | η (%) |
|--|-------------------|--|----------|---------------|
| 0 | 0 | 5.080 | - | - |
| 1×10^{-5} | | 7.832 | 0.3514 | 35.14 |
| 5×10^{-5} | | 9.236 | 0.4500 | 45.00 |
| 1×10^{-4} | | 11.080 | 0.5415 | 54.15 |
| 5×10^{-4} | | 12.594 | 0.5966 | 59.66 |
| 1×10^{-3} | | 16.634 | 0.6946 | 69.46 |
| 0 | 150 | 4.365 | - | - |
| 1×10^{-5} | | 6.145 | 0.2897 | 28.97 |
| 5×10^{-5} | | 6.741 | 0.3525 | 35.25 |
| 1×10^{-4} | | 8.105 | 0.4614 | 46.14 |
| 5×10^{-4} | | 9.167 | 0.5238 | 52.38 |
| 1×10^{-3} | | 10.988 | 0.6027 | 60.27 |
| 0 | 2000 | 2.814 | - | - |
| 1×10^{-5} | | 3.358 | 0.1620 | 16.20 |
| 5×10^{-5} | | 3.600 | 0.2183 | 21.83 |
| 1×10^{-4} | | 3.914 | 0.2810 | 28.10 |
| 5×10^{-4} | | 4.465 | 0.3698 | 36.98 |
| 1×10^{-3} | | 5.490 | 0.4874 | 48.74 |

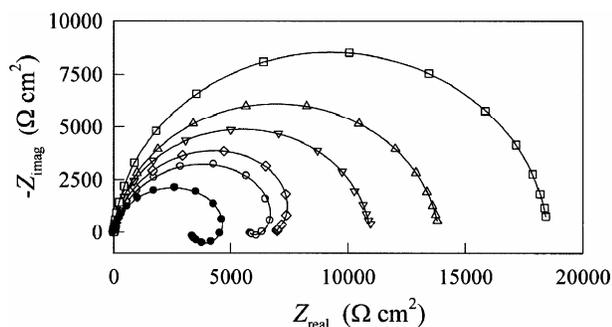


Fig. 7—The Nyquist plot of the Al-2.5Mg alloy in a 0.5 mol dm⁻³ NaCl solution at T = 20°C, without (●) and in the presence of gentisic acid in the concentration of 1×10^{-3} mol dm⁻³ (□), 5×10^{-4} mol dm⁻³ (Δ), 1×10^{-4} mol dm⁻³ (∇), 5×10^{-5} mol dm⁻³ (◇) and 1×10^{-5} mol dm⁻³ (○).

at high frequencies has often been attributed to the formation of an oxide layer¹⁸⁻²⁰ or to an oxide layer itself. Some authors attribute its existence to the transport process through the aluminium oxide layer^{21,22}. The origin of the inductive loop can be ascribed to the relaxation of adsorbed species and alloy dissolution^{20,21,23}.

The addition of gentisic acid leads to growth in diameter and size of the capacitive semicircle, which probably reflects physical blocking of the alloy surface. Furthermore, at inhibitor concentration higher than 5×10^{-5} mol dm⁻³ the inductive loop in the Nyquist complex plane completely disappears, which indicates the formation of a thicker protective film and therefore reduced corrosion.

In mathematical analysis of impedance diagrams the constant phase element, CPE, was used instead of an ideal capacitor to account for the deviations observed, as capacitive loops are depressed. The impedance of CPE is described by the expression^{24,25}:

$$Z_{CPE} = Q^{-1}(j\omega)^{-n} \quad \dots (9)$$

where Q and n are constants, that can be used in data fitting for interpreting frequency dispersions.

Figure 8 shows the equivalent circuits used to fit the experimental data. The equivalent circuit marked with (a) was found to fit the experimental data obtained in the NaCl solutions without, and with the addition of gentisic acid in concentrations 1×10^{-5} mol dm⁻³ and 5×10^{-5} mol dm⁻³. It consists of a CPE (substituting for capacity in the equivalent circuit $n \approx 1$ and $Q \approx C$) in parallel to the series resistors R_1 and R_2 and an inductance L in parallel to R_2 . R_{el} corresponds to the electrolyte resistance and it was found to be of the order of 4 Ω cm². The equivalent circuit marked (b) is used for modeling the

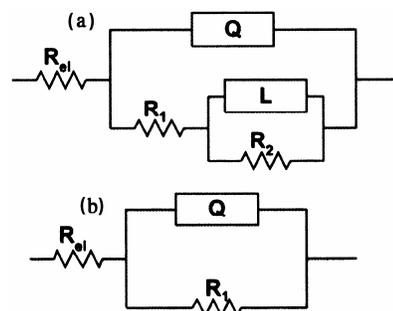


Fig. 8—Equivalent circuits for modeling impedance data of the Al-2.5Mg alloy.

Table 4—Impedance parameters for the Al-2.5Mg alloy in 0.5 mol dm⁻³ NaCl solution in the absence and in the presence of different concentrations of the gentisic acid

| C (mol dm ⁻³) | Q ₁ ×10 ⁶ (Ω ⁻¹ s ⁿ cm ⁻²) | n ₁ | R ₁ (kΩ cm ²) | L (kH cm ⁻²) | R ₂ (kΩ cm ²) |
|------------------------------|---|----------------|---|-----------------------------|---|
| 0 | 22.63 | 0.88 | 3.34 | 1.32 | 1.86 |
| 1×10 ⁻⁵ | 20.77 | 0.88 | 5.83 | 1.10 | 2.15 |
| 5×10 ⁻⁵ | 18.98 | 0.90 | 7.01 | 0.75 | 2.50 |
| 1×10 ⁻⁴ | 15.42 | 0.93 | 10.96 | | |
| 5×10 ⁻⁴ | 13.95 | 0.92 | 13.90 | | |
| 1×10 ⁻³ | 11.24 | 0.95 | 18.51 | | |

experimental results for measurements in NaCl solution with higher concentration of gentisic acid. It consists of a parallel *RQ* combination connected in series with the electrolyte resistance.

The parameters of the equivalent circuit were evaluated using the simple last squares fit procedure and are presented in Table 4. From the Table it can be seen that, when the gentisic acid concentration increases from 1×10⁻⁵ to 1×10⁻³ mol dm⁻³, the values of *R*₁ increases from 5.83 to 18.51 kΩ cm², and the values *Q* decreases from 20.77 to 11.24 Ω⁻¹ sⁿ cm⁻². The increase in *R*₁ indicates an increase in corrosion resistance whereas the decrease in *Q* values can be correlated to a decrease in the corroded area by formation of a thicker protective film and an increase in electrode area covered by inhibitor.

Gentisic acid belongs to the group of phenolic acid and has chemical structure of an aromatic nucleus, two hydroxyl groups and one carboxyl group. The literature indicates that organic compounds that contain hydroxyl or carboxyl groups may form complexes with metallic cations^{22,26-28}. The inhibitory action of gentisic acid could therefore be explained by the formation of complex molecules with aluminium ions and then precipitation of these complexes on aluminium alloy surface in places where oxide film was destroyed. Similar mechanism was proposed for corrosion inhibition of Al-2.5Mg alloy by (+)-catechine¹.

Conclusion

The results of polarization measurements have indicated that the gentisic acid acts as a cathodic inhibitor of the Al-2.5Mg alloy in a 0.5 mol dm⁻³ NaCl solution. The inhibiting efficiency of the applied inhibitor is highest in a quiescent solution and at the lowest temperature examined and decreased with increasing electrode rotation rate and increasing electrolyte temperature.

The adsorption behaviour of gentisic acid can be described by the Freundlich adsorption isotherm. The values determined for standard free adsorption energies indicate physical adsorption of gentisic acid on the electrode surface.

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