J. Mater. Environ. Sci. 6 (3) (2015) 892-901
ISSN : 2028-2508
CODEN: JMESCN

Corrosion inhibition of aluminum and aluminum silicon alloys in sodium hydroxide solutions by methyl cellulose

Salah Eid*, M. Abdallah, E. M. Kamar, A. Y. El-Etre
Chemistry department, Faculty of science, Benha University, Benha, Egypt

Received 8 May 2014; Revised 8 January 2015; Accepted 20 January 2015
**Corresponding author: eedsalah@yahoo.com ;*

Abstract
Methyl cellulose was tested as inhibitor for corrosion of aluminum and aluminum silicon alloys in 0.1M NaOH solution. The inhibition action of methyl cellulose was studied using potentiostatic polarization, electrochemical impedance spectroscopy (EIS), cyclic voltammetry and potentiodynamic anodic polarization techniques. Effect of temperature on the inhibition efficiency of studied and the values of activation thermodynamic parameters were calculated and explained. The inhibition efficiency increases with increasing the concentration of methyl cellulose and decreases with increasing temperature as well as Si content. This was attributed to a lower affinity of the inhibitor to adsorb on Si than on Al. The inhibition action was explained on the basis of adsorption of methyl cellulose on the surface of Al or Al-Si alloys forming a barrier of mass and charge transfer leading to protect the metal surface from the aggressive ions.

Key words: Aluminum - Aluminum Silicon Alloys - Methyl cellulose - Corrosion inhibitors - Adsorption.

1. Introduction
The corrosion studies of aluminum (Al) and aluminum silicon (Al-Si) alloys are closely related to their wide applications in industry. The corrosion behavior of aluminum and aluminum alloys has been studied galvanostatically and potentiodynamically in alkaline media [1-3]. Nowadays, besides the efficiency of the compound which used as inhibitor, another important factor has to be taken into account to choose the suitable inhibitor. In this sense, despite the demonstrated efficiency of chromate as inhibitor, the known toxicity and carcinogenic properties of Cr(VI) compounds exert a continuous pressure to use green inhibitors with null toxicity [4-6]. Many natural products have been previously used as corrosion inhibitors for different metals in various environments [7-13]. The aim of the present work is to study the inhibiting action of methyl cellulose on the general and pitting corrosion of Al and Al-Si in 0.1 M NaOH. The effect of Si content on the corrosion rate as well as the inhibition efficiency was also studied. Potentiostatic polarization, electrochemical impedance spectroscopy (EIS), cyclic voltammetry and potentiodynamic anodic polarization techniques were used in the study.

2. Materials and methods
The working electrodes were made of pure Al and two of Al-Si alloys. The composition and the area of the working electrodes are cited in table (1).

Table (1): Composition and area of used electrodes.

Sample	Si	Fe	Cu	Mn	Mg	Ni	Ti	Zn	Na	A, cm^2
Al	-	-	-	-	-	-	-	-	-	0.487
Alloy 1 (6063)	0.42	0.17	0.001	0.009	0.42	0.001	0.010	0.001	0.0012	0.45362
Alloy II (20556)	7.01	0.11	0.000	0.000	0.318	0.001	0.091	0.001	0.0012	0.347

Rods of the used metals were embedded in Araldite holders so as only the bottom circular areas are exposed to the corrosive medium. Before being used, the electrode was polished successively with different grades of emery papers until 2500 grad, degreased with acetone and then rinsed with distilled water. A Pt foil was used as a counter electrode. The potential was measured against a reference saturated calomel electrode (SCE). All solutions were freshly prepared using analytical grade chemicals and distilled water. The structure of methyl cellulose is given in Fig. (1). All experiments were carried out at 30 ± 1°C. Potentiostatic polarization and potentiodynamic anodic polarization experiments were carried out using a PS remote potentiostat with PS6 software, while cyclic voltammetry and Impedance were controlled by SP-150 potentiostate/galvanostate with 092-06/2 Potentio.Galvano board with EIS option at 30 ± 1°C.

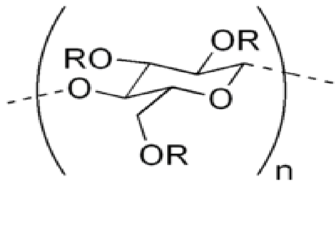

Figure (1): Methylcellulose structure

3. Results and discussion

3.1. Potentiostatic polarization

The potentiostatic polarization curves of pure aluminum in 0.1 NaOH solutions; free and inhibited with different concentrations of methyl cellulose were traced at scan rate of 10 mV/sec and shown in Fig (2). Similar curves were obtained for alloy I and alloy II (not shown). The respective electrochemical parameters e.g. current density (I_{corr}), corrosion potential (E_{corr}) anodic and cathodic Tafel slopes (β_a and β_c), the degree of surface coverage (θ) and the percentage of inhibition efficiency (%IE) are given in table (2).

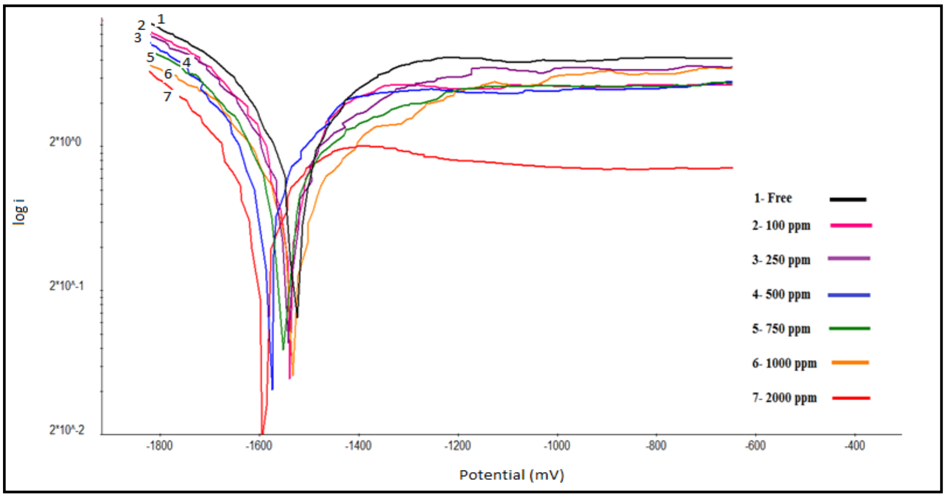

Figure (2): Anodic and cathodic polarization curves for aluminum electrode in 0.1 M NaOH solution and different concentrations of methyl cellulose at scan rate of 10 mV/sec

Inspection of this table shows that, the value of corrosion current density (I_{corr}) decreases and the inhibition efficiency (IE%) increases with increasing the concentration of the methyl cellulose, indicating the inhibiting effect of the inhibitor toward the dissolution of aluminum and its alloys in the alkaline solution. The values of corrosion potential as well as those of Tafel constants did not affected greatly upon the addition of increasing concentrations of methyl cellulose. These findings suggest that methyl cellulose act as mixed inhibitor and do not interfere with the dissolution mechanism of aluminum in the alkaline solution. This means that the methyl cellulose molecules adsorb on both anodic and cathodic sites on aluminum surface making a barrier between the metal and the corrosion medium.

The value of inhibition efficiency (IE %) decreases with increasing silicon content in the following order:

Aluminum > alloy I > alloy II

This may be attributed to a lower affinity of inhibitors to adsorb on silicon than that on aluminum. Therefore, increasing of silicon content reduces the strength of adherence of the adsorbed inhibitors molecules leading to lowering of the inhibition efficiency value.

Table (2): Corrosion parameters obtained from potentiostatic polarization measurements of aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution containing different concentrations of methyl cellulose at 30 ^{o}C.

Electrode type	Inh. conc. (ppm)	β_a mV. decade^{-1}	$-\beta_c$ mV. decade^{-1}	$-E_{corr}$ mV (SCE)	I_{corr} µA/cm^2	θ	%IE
Al	0	264	127	1545	435.4	0	0
	100	289	127	1555	379.8	12.77	0.1277
	250	269	135	1544	337.7	22.44	0.2244
	500	279	122	1593	298.4	31.47	0.3147
	750	260	130	1555	251.6	42.21	0.4221
	1000	247	130	1537	196.5	54.87	0.5487
	2000	261	114	1607	167.7	61.48	0.6148
Alloy I	0	272	122	1592	376.8	0	0
	100	269	135	1545	337.7	10.38	0.1038
	250	269	113	1602	306.3	18.71	0.1871
	500	309	115	1605	267.0	29.14	0.2914
	750	292	128	1548	232.5	38.30	0.3830
	1000	269	128	1529	187.7	50.19	0.5019
	2000	302	117	1554	160.7	57.35	0.5735
Alloy II	0	286	115	1619	272.5	0	0
	100	308	107	1642	249.2	8.55	0.0855
	250	325	116	1588	230.3	15.49	0.1549
	500	336	134	1601	207.8	23.74	0.2374
	750	331	115	1582	188.9	30.68	0.3068
	1000	323	109	1628	167.7	38.46	0.3846
	2000	273	132	1580	149.5	45.14	0.4514

3.2. Adsorption isotherm

The adsorption behavior of methyl cellulose on the Al surface and its alloys can be interpreted by finding a suitable isotherm. A number of mathematical relationships for the adsorption isotherms have been suggested to fit the experimental data of the present work. The equation that fits our results is that due to Freundlich isotherm [14] which is given by the general equation:

$$\theta = KC^n \quad (1)$$

or alternatively by:

$$\log \theta = \log K + n \log C \quad (2)$$

where K and C are the equilibrium constant of the adsorption process and additive concentration, respectively.

A plot log θ against log C gives a straight line of intercept, log K. The plot is shown in fig (3).

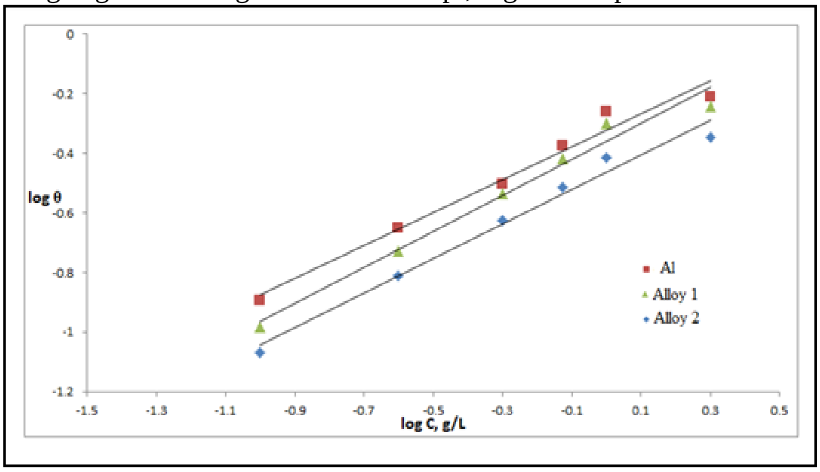

Figure (3): The relation between log (θ) and log C, g/L for aluminum, alloy I and alloy II electrode in 0.1 M NaOH solution and different concentrations of methyl cellulose.

The equilibrium constant of adsorption, K, is related to the standard free energy of adsorption, ΔG^o_{ads}, with the following equation [15]:

$$K=1/55.5 \exp (- \Delta G^o_{ads} /RT) \quad (3)$$

where T is the absolute temperature and R is the gas constant (8.314 J. $mol^{-1}.K^{-1}$). The numerical value 55.5 is the concentration of water in $mol.L^{-1}$ which will be replaced with 1000 $g.L^{-1}$ (which is the unit used to determine the additive concentration) in our calculations:

$$K=1/1000 \exp (- \Delta G^o_{ads} /RT) \quad (4)$$

The equilibrium constant and the adsorption free energy of methyl cellulose adsorbed on the surface of aluminum and its alloys in 0.1 M NaOH were given in table (3). The standard free energy change of adsorption is associated with water adsorption/desorption equilibrium which forms an important part in the overall free energy changes of adsorption. The negative value of ΔG^o_{ads} indicates that the adsorption process of these compounds on the metal surface is spontaneous one.

Table (3): Adsorption parameters of methyl cellulose on the surface of aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution.

Electrode type	K	$-\Delta G^o_{ads}$ kJ. mol^{-1}
Aluminum	0.4754	15.53
Alloy I	0.4358	15.31
Alloy II	0.3450	14.72

3.3. Effect of temperature

The effect of increasing temperature on the potentiostatatic polarization curves of aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution and in presence 2000 ppm methyl cellulose was studied at scan rate of 10 mV/sec. The increasing in temperature causes a shift of the polarization curves toward more negative direction and high current densities. The shift increases as the temperature is increased. This result indicates that the increase of temperature increases the corrosion rate of aluminum and its alloys in the alkaline solution.

Tables (4 & 5) show the electrochemical parameters of increasing temperature on the potentiostatic polarization curves of aluminum and its alloys in 0.1 M NaOH solution in the absence and presence of 2000 ppm methyl cellulose, respectively, at scan rate 10 mV/sec. It was found that, the value of inhibition efficiency (IE %) of methyl cellulose on aluminum, alloy I and alloy II electrodes in 0.1 NaOH decreases with increasing the temperature. It is obvious that increase temperature increases the rate of desorption of the additives molecules from the electrode surface which leads directly to decreasing of inhibition efficiency.

Table (4): Corrosion parameters obtained from potentiostatatic polarization measurements of aluminum alloy I and alloy II electrodes in 0.1 M NaOH solution at different temperatures.

Electrode type	T, ^oC	β_a mV.$decade^{-1}$	$-\beta_c$ mV.$decade^{-1}$	$-E_{corr}$ mV	I_{corr} $\mu A/cm^2$
Al	30	267	127	1545	435.4
	40	257	132	1547	470.0
	50	258	121	1577	554.3
	60	255	120	1588	775.2
Alloy I	30	272	122	1592	376.8
	40	328	115	1620	429.8
	50	235	141	1600	475.2
	60	234	134	1604	660.3
Alloy II	30	286	115	1619	272.5
	40	259	135	1589	317.9
	50	239	141	1603	359.3
	60	242	143	1679	456.5

Table (5): Corrosion parameters obtained from potentiostatatic polarization measurements of aluminum, alloy I and alloy II electrodes in 0.1M NaOH and 2000 ppm methyl cellulose solution at different temperature.

Electrode type	T °C	β_a mV. decade^{-1}	$-\beta_c$ mV. decade^{-1}	$-E_{corr}$ mV	I_{corr} μA/cm^2	θ	%IE
Al	30	261	114	1607	167	0.61	61
	40	332	109	1658	196	0.58	58
	50	319	131	1594	261	0.52	52
	60	245	102	1605	428	0.44	44
Alloy I	30	302	117	1554	160	0.57	57
	40	317	114	1595	190	0.55	55
	50	319	121	1574	251	0.47	47
	60	310	122	1583	373	0.43	43
Alloy II	30	273	132	1580	149	0.45	45
	40	317	115	1595	183	0.42	42
	50	298	129	1634	225	0.37	37
	60	332	130	1648	310	0.33	33

3.4. Activation energy calculations

The activation energy E_a for the corrosion of aluminum and its alloys electrodes in absence and presence of 2000 ppm methyl cellulose was calculated using Arrhenius type equation [15]:

$$\log i = \log A - (E_a/2.303RT) \quad (5)$$

where E_a is the apparent activation energy, R is the universal gas constant, A is the Arrhenius pre-exponential factor, T is the absolute temperature and i is the corrosion current obtained from tables (3&4). Plots of logarithmic corrosion current (log i) against the reciprocal of absolute temperature (1/T) are shown graphically in fig. (4). The values of the activation energies (E_a) were calculated from the slopes of the straight lines and given in Table (6).

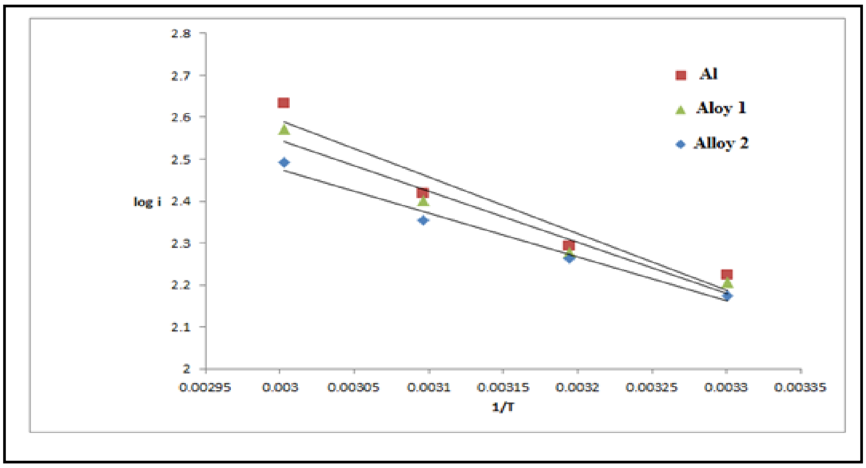

Figure (4): Arrhenius plot for aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution + 2000 ppm of methyl cellulose.

The enthalpy and entropy of activation for the corrosion of aluminum and its alloys in absence and presence of inhibitor were calculated using the transition state equation [15]:

$$\log (i/T) = [(\log R/hN) + (\Delta S^*/2.303RT)] - (\Delta H^*/2.303RT) \quad (6)$$

where, h is Planck's constant, N is Avogadro's number, ΔS^* is the entropy of activation and ΔH^* is the enthalpy of activation. Fig (5) represents the plot of log k/T against 1/T. A straight line is obtained with slopes of ($-\Delta H^*/2.303R$) and an intercept of ($\log R/Nh + \Delta S^*/2.303R$), from which the values of ΔH^* and ΔS^* were calculated, respectively. The calculated values of activation enthalpies, ΔH^* and activation entropies, ΔS^* are given in Table (6). Values of the entropy of activation ΔS^* in the absence and presence of the studied compounds are negative .This implies that the activated complex in the rate determining step represents an association rather than a dissociation step [16]. This means that the activated molecules were in higher order state than that at the initial stage [17]. The positive values of ΔH^* has reflect the exothermic nature of the corrosion process.

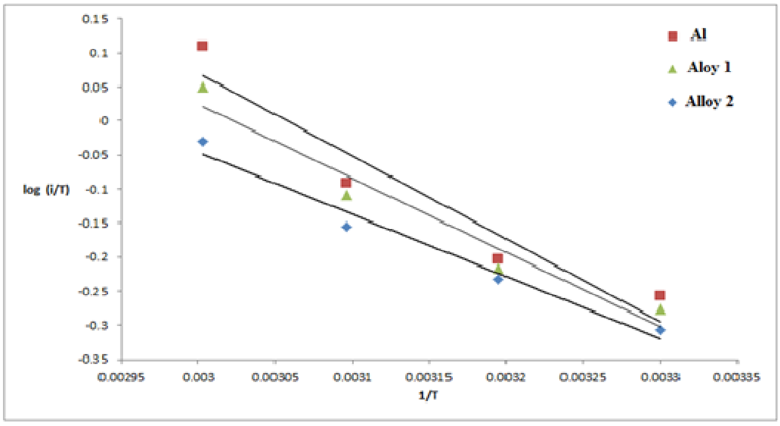

Figure (5): Plots of log (i/T) versus 1/T for aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution + 2000 ppm of methyl cellulose.

Table (6): the values of activation parameters of Aluminum alloy 6063 in 0.1 NaOH and 2000 ppm methyl cellulose.

inhibitor type	Electrode type	$-\Delta S^*$ J. mol^{-1}.K^{-1}	ΔH^* kJ. mol^{-1}	E_a kJ. mol^{-1}
Al	Free	151.79	13.12	15.75
	Methyl cellulose	126.64	23.20	25.84
Alloy I	Free	155.79	12.21	14.85
	Methyl cellulose	134.72	20.80	23.44
Alloy II	Free	161.24	11.21	13.96
	Methyl cellulose	146.03	17.48	20.11

3.5. Electrochemical Impedance Spectroscopy (EIS)

Fig (6) represents the effect of addition of increasing concentrations of methyl cellulose on the Nyquist plots curves of alloy II electrode in 0.1 M NaOH solution. Inspection of the figure reveals that each impedance diagram consists of a large capacitive loop at high frequency and a small inductive one at low frequency values. The capacitive loop could be assigned to the relaxation process in the natural oxide film covering the surface of the electrode and its dielectric properties [18]. The oxide film is considered to be a parallel circuit of a resistor due to the ionic conduction in the oxide film, and a capacitor due to its dielectric properties. The inductive loop may be related to the relaxation process obtained by adsorption and penetration of OH^- ions on and into the oxide film [19].

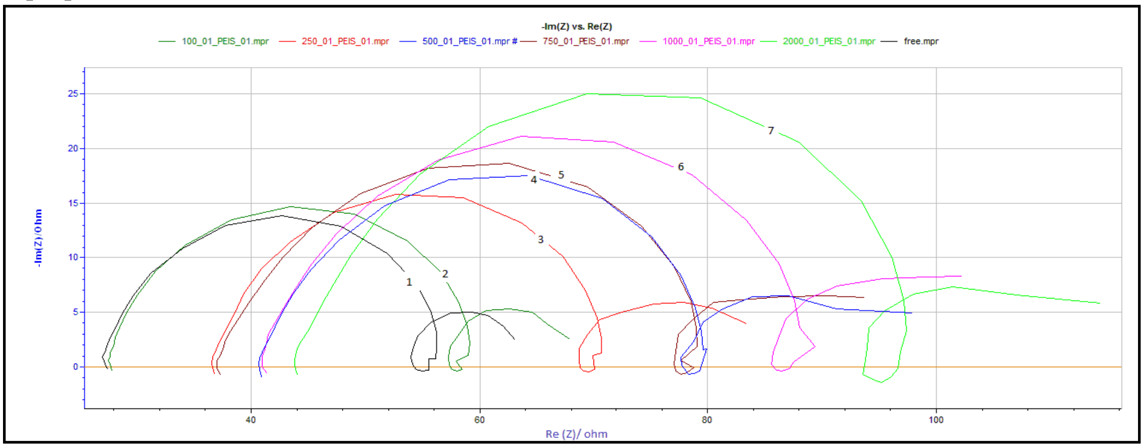

Figure (6): Nyquist plots recorded for alloy II electrode in 0.1 M NaOH solution and different concentrations of methyl cellulose at 30°C.

ISSN : 2028-2508
CODEN: JMESCN

It is essential to develop the appropriate models for the impedance which can then be used to fit the experimental data and extract the parameters which characterize the corrosion process. The equivalent circuit model used to fit the experimental data is shown in fig (7) similar to those found in literature [20-21]. In this circuit, R_{ct} is the charge transfer resistance, C_{dl} is the double layer capacitance, R_t the interfacial charge-transfer resistance, L the inductance, R_L the inductive resistance and R_p the polarization resistance.

Table (7) shows the effect of increasing concentrations of NaOH on some electrochemical kinetic parameters e.g charge transfer resistance (R_{ct}), the capacity of double layer (C_{dl}), the degree of surface coverage (θ) and the percentage of inhibition efficiency (%IE) during impedance measurement of Aluminum. It can see from this Table that the values of charge transfer resistance (R_{ct}) increase with increasing inhibitor concentration. This increase in R_{ct} with inhibitor concentration may be result of more inhibitor molecules adsorption on the metal surface and decreasing of C_{dl} may be caused by reduction in local dielectric constant and /or by increase in the thickness of the electrical double layer. This result suggests that the inhibitor molecules act by adsorption at the metal/ solution interface [22].

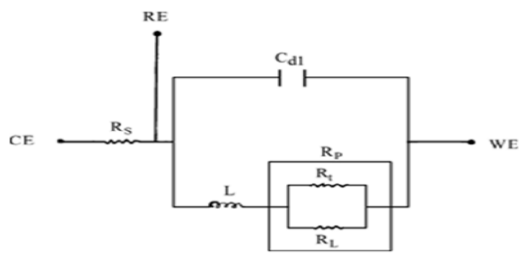

Figure (7): The equivalent circuit model used to fit the experimental data.

Table (7): Electrochemical impedance parameters obtained by EIS technique for aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution and different concentrations of methyl cellulose at 30˚C.

Electrode type	Inh. Conc. (ppm)	R_{ct}, Ω cm^2	C_{dl}, F cm^{-2}	IE%
Al	0	4.8	14.98×10^{-3}	0
	100	5.5	12.30×10^{-3}	12.73
	250	6.2	10.4×10^{-3}	22.58
	500	7.1	7.68×10^{-3}	32.39
	750	8.5	5.31×10^{-3}	43.53
	1000	10.5	3.16×10^{-3}	54.29
	2000	12.6	2.28×10^{-3}	61.91
Alloy I	0	6.7	7.09×10^{-3}	0
	100	7.5	7.52×10^{-3}	10.67
	250	8.3	4.85×10^{-3}	19.28
	500	9.7	3.92×10^{-3}	30.93
	750	11.1	2.96×10^{-3}	39.27
	1000	13.2	2.02×10^{-3}	49.24
	2000	15.6	1.44×10^{-3}	57.05
Alloy II	0	10.0	3.31×10^{-3}	0
	100	10.9	2.86×10^{-3}	8.26
	250	11.8	2.45×10^{-3}	15.25
	500	13.4	1.95×10^{-3}	24.81
	750	14.6	1.68×10^{-3}	31.51
	1000	16.1	1.34×10^{-3}	37.89
	2000	18.3	1.00×10^{-3}	45.36

3.6. Cyclic voltammetry

Fig (8) represents the effect of addition of increasing concentrations of methyl cellulose on the cyclic voltammetry curves of aluminum electrode in 0.1 M NaOH solution at scan rate 1 V/sec. Inspection of fig (8) reveal that, there is one large anodic peak in the oxidation half cycle without presence of any cathodic peaks in the reduction one. The peak potential shifts to more positive values and peak current increases as the concentration of NaOH increase.

Table (8) shows the effect of increasing concentrations of methyl cellulose on the electric charge, the degree of surface coverage (θ) and the percentage of inhibition efficiency (%IE) during the corrosion aluminum and its alloys in 0.1 M NaOH solution.

It was found that, the value of electric charge decreases and the inhibition efficiency (%IE) increases with increasing the concentration of the inhibitor, indicating the inhibiting effect of this compound toward the dissolution of aluminum and its alloys in 0.1 M NaOH.

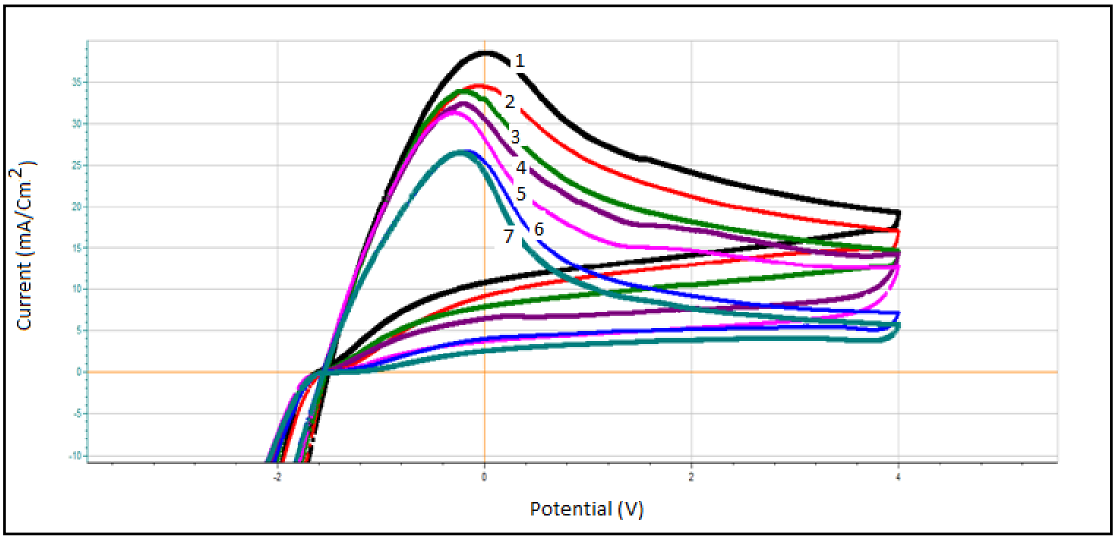

Figure (8): Cyclic voltammetry curves for aluminum electrode in 0.1 M NaOH solution and different concentrations of methyl cellulose at scan rate of 1 V/sec.

Table (8): Electric charge, Q, obtained from cyclic voltammetry measurements of aluminum, alloy I and alloy II electrodes in 0.1 M NaOH solution containing different concentrations of methyl cellulose at 30 ^oC.

Electrode type	Inh. Conc. (ppm)	Q mC/cm^2	IE%
Al	0	206.1	0
	100	180.6	12.37
	250	162.4	21.20
	500	142.6	30.81
	750	118.3	42.60
	1000	92.8	54.97
	2000	79.9	61.20
Alloy I	0	193.1	0
	100	173.4	10.20
	250	155.2	19.60
	500	135.1	30.03
	750	114.5	40.70
	1000	98.5	49.02
	2000	82.2	57.41
Alloy II	0	189.4	0
	100	174.4	7.92
	250	160.8	15.10
	500	142.2	24.92
	750	132.2	30.20
	1000	117.9	37.75
	2000	101.2	46.57

3.7. Pitting corrosion

The effect of the concentrations of methyl cellulose on the potentiodynamic anodic polarization of alloy I in [0.1 M NaOH + 0.1 M NaCl] at scan rate of 1.0mV/s is illustrated in fig (9). Similar curves were obtained for aluminum and alloy II (not shown). It was found that increasing the concentration of methyl cellulose causes a shift of the pitting potential into noble direction indicating an increased resistance to pitting attack. Straight line relationship between E_{pit} and log $C_{inh.}$, fig (10), was obtained.

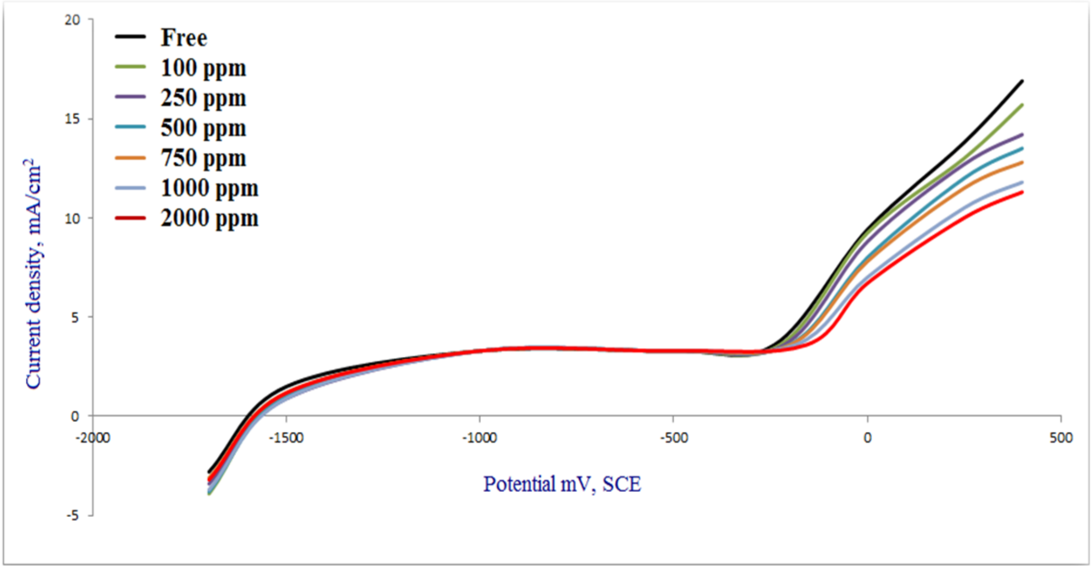

Figure (9): Potentiodynamic anodic polarization curves for alloy I electrode in 0.1 M NaOH + 0.1 M NaCl solutionand different concentrations of methyl cellulose at scan rate of 1.0 mV/sec.

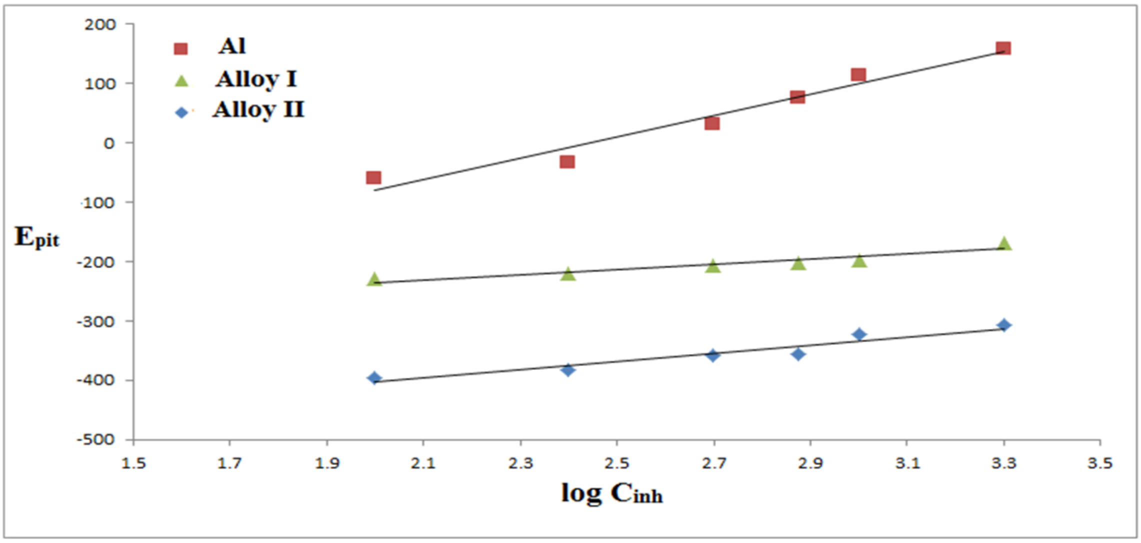

Figure (10): The relation between the pitting potential (E_{pit}) and log C_{inh}

3.8. Mechanism of Inhibition

In view of these results it is clear that the majority of organic compound containing oxygen atoms which have a pair of free electrons and, so this compound will be able to adsorb on aluminum or Al-Si alloys surface through the free electrons on the oxygen atom The occurrence of the adsorption of the compound on aluminum or Al-Si alloys surface constitutes the barrier for mass and charge transfer, leading to protect the metal surface from the attack of the aggressive anions.

Conclusions

- The results obtained from all electrochemical measurements showed that the inhibition efficiency increases with increasing inhibitor concentration.
- The values of inhibition efficiencies obtained from the different techniques used showed an agreement of the results.
- The adsorption of methyl cellulose on aluminum and Al-Si alloys surfaces in NaOH solution follows Freundlich isotherm.
- The negative values of $\Delta G°ads$ show the spontaneity of the adsorption process.
- The methyl cellulose is adsorbed on aluminum or Al-Si alloys surface forming a barrier film and protect aluminum or Al-Si alloys substrate against corrosion in 0.1 M NaOH solution.

References

1. Garrigues L., Pebere N. and Dabosi F., *Electrochim. Acta*, 41 (1996) 1209
2. Diggle J. W., Downie T.C. and Goulding C.W., *Electrochim. Acta*, 15(7) (1970) 1079
3. Baumgaertner M., and Kaesche H., *Corros. Sci*., 31 (1990) 231
4. Quraishi M. A., Ambrish Singh, Singh V. K., Yadav D. K., Singh A. K., *Mater. Chem. Phys*., 122 (2010) 114
5. Singh A., Singh A V. K. and Quraishi M. A., *Rasayan J. Chem.*, 3 (2010) 811
6. Singh A., Ahamad I., Singh V. K. and Quraishi M. A., *Chem. Engg. Communication*, 199 (2012) 63
7. El-Etre A. Y., *Corros. Sci*., 40 (11) (1998) 1845
8. Zhang S. S., and Jow T.R., *J. Power Sources*, 109(2) (2002) 422
9. Abdallah M., Zaafarany I., Al-Karanee S. O. and Abd Elfattah, *Arab. J. Chem.*, 5(2) (2012) 225
10. Gunasekaran G. and Chauhan L.R., *Electrochim. Acta* 49(25) (2004) 4387
11. El-Etre A.Y., Abdallah M. and El-Tantawy Z.E., *Corros. Sci.*, 47(2) (2005) 385
12. Abdel-Gaber A. M., Abd-El-Nabey B. A., Sidahmed I. M., El-Zayady A. M., Saadawy M., *Corros. Sci.* 48(9) (2006) 2765
13. Oguzie E. E, *Corros. Sci*., 49(3) (2007) 1527
14. Fouda A. S, Elewady G. Y., El-askalany A. and Shalabi K., *ZAŠTITA MATERIJALA* 51(4) (2010) 205
15. Ostovari A., Hoseinieh S. M., Peikari M., Shadizadeh S. R. and Hashemi S. J., Corros. *Sci*., 51(9) (2009) 1935
16. Bouklah M., Benchat N., Hammouti B. Aouniti A. and Kertit S., *Mater. Lett.*, 60 (2006) 1901
17. Fouda A.S., Al-Sarawy A. A. and El-Katori E. E., *Desalination*,201(2006) 1
18. Mansfeld F., Lin S., Kim S. and Shih H., *Corros*. *Sci*., 27(9)(1987) 997
19. Lenderink H. J. W., Linden M. V. D. and De Wit J. H. W., *Electrochim*. *Acta*, 38(1993) 1989
20. Brett C. M. A, *Corros. Sci.*, 33(2) (1992) 203.
21. Abdel-Rehim S. S., Hassan H. H. and Amin M. A., *Appl*. *Surf*. *Sci*., 187 (2002) 279.
22. Lagrenee M., Mernari B., Bouanis B., Traisneland M. and Bentiss F., *Corros*. *Sci*., 44 (3) (2002) 573