Influence of zinc sulphate on the corrosion resistance of L80 alloy immersed in sea water in the absence and presence of sodium potassium tartrate and trisodium citrate

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Abstract

Seawater can be used in cooling water systems. L80 can be used as pipeline carrying sea water. However, this alloy will undergo corrosion. Corrosion can be prevented by addition of inhibitors such as sodium potassium tartrate (SPT), trisodium citrate (TSC), and zinc sulphate. Corrosion resistance of L80 alloy in sea water in the absence and presence of the above inhibitors has been evaluated by polarisation study and AC impedance spectra. It is observed that SPT and TSC show better inhibition efficiency in the presence of Zn^{2+} . Further it is found that SPT–Zn system is better than the TSC–Zn system. When the SPT or TSC inhibitor is added to sea water, a protective film is formed on the metal surface. This prevents the transfer or release of electrons from the metal surface to the bulk of the solution. Thus corrosion is controlled. This is revealed by the fact that, during polarisation study, the linear polarisation resistance value increases and corrosion current value decreases. During electrochemical impedance study, charge transfer value increases and double layer capacitance value decreases. In the presence of Zn^{2+} , corrosion resistance of the metal further increases, which is supported by the fact that the linear polarisation resistance value further increases and corrosion current value further decreases. Similarly during electrochemical impedance study, the charge transfer resistance increases tremendously and the double layer capacitance value decreases to a great extent. It is inferred that in the presence of Zn^{2+} , more inhibitor is transported towards the metal surface as a Zn^{2+} –inhibitor complex. On the metal surface, an iron inhibitor complex is formed on the anodic sites of the metal surface and Zn^{2+} is released. The released Zn^{2+} combines with OH^- to form $\text{Zn}(\text{OH})_2$ on the cathodic sites of the metal surface. Thus in the presence of Zn^{2+} , both anodic reaction and cathodic reaction are controlled effectively. This accounts for the increase in corrosion resistance of metal in sea water in the presence of inhibitor and Zn^{2+} .

Keywords: corrosion inhibition, seawater, sodium potassium tartrate, trisodium citrate, polarisation study, AC impedance spectra, L80 alloy.

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Introduction

Seawater makes up the oceans and seas, covering more than 70 percent of Earth's surface. Seawater is a complex mixture of 96.5 percent water, 2.5 percent salts, and smaller amounts of other substances, including dissolved inorganic and organic materials, particulates, and a few atmospheric gases [1]. Almost anything can be found in seawater. This includes dissolved materials from Earth's crust as well as materials released from organisms. The most important components of seawater that influence life forms are salinity, temperature, dissolved gases (mostly oxygen and carbon dioxide) and nutrients [2].

Seawater can be used in cooling water systems, especially in ships and in marine environments. In these systems L80 alloy can be used to carry out the seawater. Hence knowledge of corrosion resistance of L80 alloy in seawater in presence of inhibitors will be useful. Corrosion resistance of aluminium and its alloys [3, 4], zinc and its alloys [5, 6], copper and its alloys [7, 8], mild steel [9, 10], stainless steel [11, 12], nickel and its alloys [13, 14] in seawater has been evaluated. Corrosion inhibitors such as, inorganic compounds [15, 16] and natural products [17, 18] have been used to prevent the corrosion of metals and alloys. In the present study corrosion resistance of L80 alloy in seawater in presence of sodium potassium tartrate (SPT) and trisodium citrate (TSC) has been evaluated by electrochemical studies such as polarization study and AC impedance spectra. Influence of Zn^{2+} on the corrosion resistance of the above systems has also been evaluated.

Experimental

Electrochemical studies

In the present work corrosion resistance of L80 alloy immersed in various test solutions were measured by polarization study and AC impedance spectra.

Polarization study

In the present study polarization studies were carried out in a CHI Electrochemical work station/analyzer, model 660A. It was provided with automatic IR compensation facility. A three electrode cell assembly was used (Figure 1).

The working electrode was L80 alloy. A SCE was the reference electrode. Platinum was the counter electrode. A time interval of 5 to 10 min was given for the system to attain a steady state open circuit potential. The working electrode and the platinum electrode were immersed in sea water in the absence and presence of inhibitor. Saturated calomel electrode was connected with the test solution through a salt bridge. From polarization study, corrosion parameters such as corrosion potential (E_{corr}), corrosion current (I_{corr}), Tafel slopes anodic =

b_a and cathodic = b_c and LPR (linear polarisation resistance) value were determined. The scan rate was 0.01 V/s. The hold time at (E_{fcs}) was zero and the quiet time was 2 seconds.

Figure 1. Three electrode cell assembly.

AC Impedance spectra

In the present investigation the same instrument and set-up used for polarization study was used to record AC impedance spectra also. A time interval of 5 to 10 min was given for the system to attain a steady state open circuit potential. The real part (Z') and imaginary part (Z'') of the cell impedance were measured in ohms at various frequencies. AC impedance spectra were recorded with initial $E(V)=0$, high frequency ($\text{Hz}=1 \cdot 10^5$), low frequency ($\text{Hz}=1$), amplitude (V)=0.005 and quiet time (s)=2. From Nyquist plot the values of charge transfer resistance (R_t) and the double layer capacitance (C_{dl}) were calculated.

$$R_t = (R_s + R_t) - R_s$$

Where R_s = solution resistance.

C_{dl} values were calculated using the relationship

$$C_{dl} = 1/2 \cdot 3.14 \cdot R_t \cdot f_{\max}$$

Where $f_{\max}$ = frequency at maximum imaginary impedance.

Sea water

The composition of sea water used in this study is given in Table 1. Sea water was collected in Bay of Bengal, located at Kanampadi, East Coast Road, Chennai, India.

Table 1. Composition of sea water.

Physical Examination	Acceptable Limit	Permissible limit	Sample Value
1. Colour	–	–	Colourless & Clear
2. Odour	Unobjectionable		Unobjectionable
3. Turbidity NT Units	1	5	0.2
4. Total dissolved solids mg/L	500	2000	29400
5. Electrical conductivity $\mu\text{Ohm}^{-1}/\text{cm}$	–	–	42000
Chemical Examination	Acceptable Limit	Permissible limit	Sample Value
6. pH	6.5–8.5	6.5–8.5	7.46
7. pH Alkalinity as CaCO_3	–	0	0
8. Total alkalinity as CaCO_3	200	600	140
9. Total hardness as CaCO_3	200	600	4000
10. Calcium as Ca	75	200	1200
11. Magnesium as Mg	30	100	240
12. Iron as Fe	0.1	1	0
13. Manganese as Mn	0.1	0.3	NT
14. Free ammonia as NH_3	0.5	0.5	0.48
15. Nitrite as NO_2	0.5	0.5	0.104
16. Nitrate as NO_3	45	45	25
17. Chloride as Cl	250	1000	15000
18. Fluoride as F	1	1.5	1.8
19. Sulphate as SO_4	200	400	1170
20. Phosphate as PO_4	0.5	0.5	1.47
21. Tids test 4 hrs as O_2	–	–	NT

L80 Alloy

Manufactured to API specification 5CT. This is a controlled yield strength material with a hardness testing requirement. L80 is usually used in wells with sour (hydrogen sulfide) environments. Chemical composition of L80 alloy is given in Table 2.

Table 2. Chemical composition of L80 alloy.

	C	Mn	Mo	Cr	Ni	Cu	Ti	P	S	Si	V	Al
Min	–	–	–	–	–	–	–	–	–	–	–	–
Max	0.430	1.900	–	–	0.250	0.350	–	0.030	0.030	0.450	–	–

The remaining Wt% was iron.

Inhibitors used

Pure samples of sodium potassium tartrate (SPT), trisodium citrate (TSC) and zinc sulphate were used as corrosion inhibitors.

Results and Discussion

Electrochemical studies

In the present work corrosion resistance of L80 alloy immersed in various test solutions were measured by polarization study and AC impedance spectra.

L80 alloy–sodium potassium tartrate (SPT) system

Polarization study

The polarization parameters are given in Table 3. The polarization curves are shown in Figures 2–4. It is seen from Table 3 that when SPT is added to sea water, the corrosion resistance of L80 alloy increases. This is revealed by the fact that when SPT is added to sea water, the *LPR* values increases and *I*_{corr} value decreases (Figures 2–5).

Table 3. Corrosion parameters of L80 alloy immersed in sea water in the absence and presence of SPT and zinc sulphate (ZnSO₄·7H₂O) obtained by polarization study.

System	<i>E</i> _{corr} mV SCE	<i>b</i> _c mV/decade	<i>b</i> _a mV/decade	<i>LPR</i> Ohm·cm ²	<i>I</i> _{corr} A/cm ²
Sea water	–830	161	221	355	1.138·10 ^{–4}
Sea water + SPT (300 ppm)	–822	119	212	677	4.908·10 ^{–5}
Sea water + SPT (300 ppm) + ZnSO ₄ (100 ppm)	–537	209	252	2.388·10 ⁷	2.079·10 ^{–9}

Figure 2. Polarisation curve of L80 alloy immersed in sea water.

Figure 3. Polarisation curve of L80 alloy immersed in sea water + SPT.

Figure 4. Polarisation curve of L80 alloy immersed in sea water + SPT + zinc sulphate.

Influence of zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$)

When $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ is added to the above system, the LPR value increases tremendously to a very great extent Figure 5 and the I_{corr} decreases very much. This indicates that in presence of SPT– $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ system the corrosion resistance of L80 alloy in sea water increases to a great extent. A protective film is formed on the metal surface. This protective film is compact and very stable. This prevents transfer of electrons from anodic site to cathodic site. Hence LPR value increases which results in decrease in corrosion current value. In addition a synergistic effect exists between SPT and Zn^{2+} ions.

Figure 5. Comparison of LPR values.

AC impedance spectra

The AC impedance spectra (Nyquist plots, Bode plots) of L80 alloy immersed in various test solutions are shown in Figures 6–11. The corrosion parameters are given in Table 4. It is observed from Table 4, that when SPT is added to sea water, the corrosion resistance of L80 alloy increases. This is revealed by the fact that there is increase in R_t value, impedance value and decrease in C_{dl} value.

Table 4. Corrosion parameters of L80 alloy immersed in sea water in the absence and presence of SPT and Zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$) obtained by AC impedance spectra.

System	R_t Ohm·cm ²	C_{dl} F/cm ²	Impedance Log(z/Ohm)	Phase angle (degree)
Sea water	5.026	0.010×10^{-4}	0.9149	22.03
Sea water + SPT (300 ppm)	5.653	9.021×10^{-7}	0.9604	24.01
Sea water + SPT (300 ppm) + ZnSO ₄ (100 ppm)	$1.281 \cdot 10^7$	$3.982 \cdot 10^{-13}$	7.123	96.33

Figure 6. AC impedance spectra (Nyquist plot) of L80 alloy immersed in sea water.

Figure 7. AC impedance spectra (Bode plot) of L80 alloy immersed in sea water.

Figure 8. AC impedance spectra (Nyquist plot) of L80 alloy immersed in sea water + SPT.

Figure 9. AC impedance spectra (Bode plot) of L80 alloy immersed in sea water + SPT.

Figure 10. AC impedance spectra (Nyquist plot) of L80 alloy immersed in sea water + SPT + zinc sulphate.

Figure 11. AC impedance spectra (Bode plot) of mild steel immersed in sea water + SPT + ZnSO_4 .

Influence of zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$)

When $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ is added to the above SPT system, the corrosion resistance further increases (Figure 12). This is revealed by the fact that there is increase in R_t value and impedance value. There is decrease in C_{dl} value. The tremendous increase in R_t value is due to the synergistic effect existing between TSC and Zn^{2+} . Thus electrochemical studies reveal that the corrosion resistance of L80 alloy in sea water decreases in the following order:

$$\text{Sea water} + \text{SPT} + \text{ZnSO}_4 > \text{Sea water} + \text{SPT} > \text{Sea water}$$

Figure 12. Comparison of R_t values.

L80 Alloy–TSC system

Polarization study

The polarization parameters of L80 alloy immersed in sea water in presence of TSC system are given in Table 5. The polarization curves are shown in Figures 13 and 14. It is seen from Table 5 that when TSC is added to sea water, the corrosion resistance of L80 alloy increases. This is revealed by the fact that when TSC is added to sea water, the LPR value increases and I_{corr} value decreases (Figures 13, 14). There is formation of a protective film on the metal surface. So transfer of electron from anodic site to cathodic site is restricted. Hence LPR value increases and correspondingly corrosion current value decreases.

Table 5. Corrosion parameters of L80 alloy immersed in sea water in the absence and presence of TSC and zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$) obtained by polarization study.

System	E_{corr} mV SCE	b_c mV/decade	b_a mV/decade	LPR $\text{Ohm} \cdot \text{cm}^2$	I_{corr} A/cm^2
Sea water	–830	161	221	355	$1.138 \cdot 10^{-4}$
Sea water +TSC (300 ppm)	–761	165	232	391	$1.076 \cdot 10^{-4}$
Sea water +TSC (300 ppm) + ZnSO ₄ (100 ppm)	–704	146	209	465	$8.044 \cdot 10^{-5}$

Figure 13. Polarisation curve of L80 alloy immersed in sea water + TSC.

Figure 14. Polarisation curve of L80 alloy immersed in sea water + TSC + zinc sulphate.

Influence of zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$)

When $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ is added to the above system, the LPR value increases tremendously to a very great extent (Figure 15) and the I_{corr} decreases very much. This indicates that in presence of SPT– $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ system the corrosion resistance of L80 alloy in sea water increases to a great extent. This is due to the synergistic effect existing between SPT and Zn^{2+} .

Figure 15. Comparison LPR values.

AC impedance spectra

The AC impedance spectra (Nyquist plots, Bode plots) of L80 alloy immersed in various test solutions are shown in Figures 16–19. The corrosion parameters are given in Table 6. It is observed from Table 6 that when TSC is added to sea water, the corrosion resistance of L80 alloy increases. This is revealed by the fact that there is increase in R_t value, impedance value and decrease in C_{dl} value.

Table 6. Corrosion parameters of L80 alloy immersed in sea water in the absence and presence of TSC and Zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$) obtained by AC impedance spectra.

System	R_t $\text{Ohm} \cdot \text{cm}^2$	C_{dl} F/cm^2	Impedance $\text{Log}(z/\text{Ohm})$	Phase angle degree
Sea water	5.026	$0.010 \cdot 10^{-4}$	0.9149	22.03
Sea water + TSC (300 ppm)	6.282	$0.022 \cdot 10^{-4}$	0.9671	26.22
Sea water + TSC (300 ppm) + ZnSO ₄ (100 ppm)	7.077	$7.206 \cdot 10^{-7}$	1.021	28.84

Figure 16. AC Impedance spectra (Nyquist plot) of L80 alloy immersed in sea water + TSC.

Figure 17. AC impedance spectra (Bode plot) of L80 alloy immersed in sea water + TSC.

Figure 18. AC impedance spectra (Nyquist plot) of L80 alloy immersed in sea water + TSC + ZnSO₄.

Figure 19. AC impedance spectra (Bode plot) of L80 alloy immersed in sea water + TSC + ZnSO₄.

Influence of zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$)

When $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ is added to the above TSC system, the corrosion resistance further increases. This revealed by the fact that there is increase in R_t values (Figure 20) and impedance value. There is decrease in C_{dl} . Thus electrochemical studies such as polarisation

study and AC impedance spectra [19–36] reveal that the corrosion resistance of L80 alloy in sea water decreases in the following order:

$$\text{Sea water} + \text{TSC} + \text{ZnSO}_4 > \text{Sea water} + \text{TSC} > \text{Sea water}$$

Figure 20. Comparison of R_t values.

Equivalent circuit diagram for the Nyquist plots

The Nyquist plots (Figures 6, 8, 10, 16 and 18) in the present study reveal that diffusion controlled reactions are involved in the corrosion controlling processes. The equivalent circuit diagram is shown is Figure 21.

Figure 21. Equivalent circuit diagram for diffusion controlled process.

Mechanism of corrosion inhibition

In presence of Zn^{2+} , corrosion resistance of the metal further increases (better than the SPT/TSC system alone), which is supported by the fact that the linear polarisation resistance value further increases and corrosion current value further decreases. Similarly during electrochemical impedance study, charge transfer value increases tremendously and double layer capacitance value decreases to a great extent. It is inferred that in presence of Zn^{2+} , more inhibitor (SPT/TSC) is transported towards the metal surface, as Zn^{2+} –inhibitor complex. On the metal surface iron inhibitor complex is formed on the anodic sites of the metal surface and Zn^{2+} is released. The released Zn^{2+} combines with OH^- to form $\text{Zn}(\text{OH})_2$ complex on the cathodic sites of the metal surface. Thus in presence of Zn^{2+} , both anodic reaction and cathodic reaction are controlled effectively. This accounts for the increase in corrosion resistance of metal in sea water in presence of inhibitor and Zn^{2+} . In presence of Zn^{2+} and inhibitor a loose complex is formed between Zn^{2+} and inhibitor. The bond between them is strong enough to carry the inhibitor to the metal surface and weak enough to release the inhibitor in presence of Fe^{2+} to form Fe^{2+} –inhibitor complex on the anodic sites of the metal surface (Figures 22 and 23).

Figure 22. Anodic and cathodic reactions in corrosion process.

Figure 23. Corrosion inhibition mechanism.

Conclusion

The corrosion resistance of L80 alloy in sea water in the absence and presence of sodium potassium tartrate (SPT), trisodium citrate (TSC) and zinc sulphate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$) has been evaluated by electrochemical studies such as polarization study and AC impedance spectra. The study leads to the following conclusions:

L80 alloy – SPT system

Electrochemical studies reveal that the corrosion resistance of L80 alloy in sea water decreases in the following order:

L80 alloy – TSC system

Electrochemical studies reveal that the corrosion resistance of L80 alloy in sea water decreases in the following order:

Addition of zinc sulphate improves the corrosion resistance of the TSC and SPT systems.

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