Research Article

EFFECT OF SODIUM TUNGSTATE AS A GREEN CORROSION INHIBITOR ON THE PASSIVATION BEHAVIOR OF MILD STEEL SHEET IN AGGRESSIVE MEDIA

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Abstract

The effect of sodium tungstate on the passivation behavior of grille sheet made by mild steel was studied using corrosion tests and electrochemical measurements in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions, open to air at 25 °C. The grille sheet showed the highest corrosion resistance properties in 1 M NaOH solution as compared with 1 M HCl and 0.5 M NaCl solutions. The corrosion resistance properties of the steel sheet was decreased with increasing the concentration of sodium tungstate up to 800 ppm and its corrosion inhibition efficiency was increased with increasing the concentration. The open circuit potential of the mild steel sheet was more negative value in 0.5 M NaCl than that in 1 M HCl, whereas more positive potential value was observed in 1 M NaOH than in 0.5 M NaCl solution. It was found that the mild steel sheet used in the study was found to be more passive in 1 M NaOH than in 1 M HCl and 0.5 M NaCl solutions. Hence, a more stable anodic passive film was formed on the surface of the steel sheet in 1 M NaOH than those in 1 M HCl and 0.5 M NaCl solutions from electrochemical measurements.

Key words: Anodic current; corrosion rate; mild steel; polarization; corrosion potential; 0.5 M NaCl solution.

Introduction

Corrosion is an undesirable phenomenon which destroys the properties of metallic materials and shortens their life (Bhattarai, 2010). It is mainly due to the spontaneous instability of the metallic substances that results from the charge−transfer reactions at interfaces between the metallic material and its environment (Bockris *et al.*, 2000). The control of such undesirable corrosion process is a subject of scientific and technological significances. However, the corrosion control techniques for a particular system is very difficult works for corrosion scientists and technologists.

Mild steel is one of the widely used structural materials, although it shows poor corrosion resistance behavior at ambient temperature. The use of inhibitors, cathodic protection methods or/and coatings are main corrosion control techniques for the mild steels. Nowadays, the corrosion control method of metallic materials using various types of eco-friendly green corrosion inhibitors is becoming a fundamental academic and research concerns of corrosion scientists and engineers (Bhattarai, 2010; Revie and Uhlig, 2008). The corrosion inhibitor is a chemical substance that when added in small amounts to a corrosive environment, effectively decreases the corrosion rate of the materials exposed to the corrosive environments (Revie and

Uhlig, 2008; Hackerman and Snaveley, 1984). Very low concentrations of chemical species with special characteristics that can intervene the corrosion kinetics and thereby control the materials corrosion is generally term as corrosion inhibitor. The use of the corrosion inhibitor to retard the detereoration of the materials is becoming one of the widely used corrosion control methods.

In this context, the use of appropriate corrosion control methods to prevent the corrosion of the mild steel requires awareness and co-operation of the entire design team, including engineers, corrosion scientists and technologists. In order to ensure the more durability of the new steel-structures, we need to understand what can be done to reduce the corrosion risk of the steel in different corrosive environments. In general, the degree of corrosion resistance of the mild steel depends on the environments; the most harmful factors are oxidizing acids and chloride ion, and the most beneficial environment is alkaline (Bhattarai, 2010; Revie and Uhlig, 2008). Steels passivate generally at a pH higher than 10, even though the passivity decreases in very strong alkaline solutions (i.e. > 13.5 pH) at high temperature where the iron has a tendency to dissolve as hypoferrite, $HFeO_2^-$ (Bhattarai, 2010; Revie and Uhlig, 2008). Therefore, a strong alkaline (not very strong) solution is generally employed in many chemical processing industries

where iron or steel equipments are used (Chawla and Gupta, 1993). Many studies about the corrosion of steels were performed using different surface analysis techniques since 1970s (Asami *et al.,* 1978; Fujimoto and Tsuchiya, 2007, Shibata, 2007). The corrosion behavior of the mild steel was mainly investigated in acids (Ameh, 2014; Amin *et al.,* 2011; Al-Turkustani, 2010; Doner *et al.,* 2012; Ebenso *et al.* 2010; Fouda *et al.,* 2013; Kivisakk, 2003; Patel *et al.,* 2009), alkalis (Gonzalez *et al.,* 2007; Newman, 1981) and NaCl solutions (Isaacs *et al.,* 2007; Reffass *et al.,* 2006; Zhou *et al.,* 2012).

Practical criteria for the selection of corrosion inhibitors from the variety of inorganic and organic compounds are not only their inhibition efficiency but also safety of use, economic constraints and compatibility with other chemicals in the system. In the past, chromates were accepted as an effective corrosion inhibitor that can passivate many metals and their alloys by forming a mono-atomic or polyatomic protective passive film on the materials surface (Bhattarai, 2010; Revie and Uhlig, 2008). However, the main disadvantage of the use of chromates is its toxicity at hexavalent chromium ions and hence in recent years, the reason for the search of less toxic corrosion inhibitors alternatives (Tayler and Chambers, 2009). Similalrly, tight restrictions of the uses and disposal of chromium ions in our surroundings by environmental regulations, there is a great interest in replacing harmful chromates inhibitor by an effective non-hazardous green corrosion inhibitors.

Last two decades, extensive researchs have led to the discovery of new class of green corrosion inhibitors those are generally environmentally freindly. Molybdates, tungstates and metavenadates so on are now being increasingly used as the green inorganic corrosion inhibitors especially because of their lower toxicity (Ali *et al.,* 2009; Celeste and Idalina, 2004; Li *et al.,* 2011; Sribharathy and Rajendran, 2012; Zhao and Zuo, 2002). It was reported that the synergistic effect of anodic inhibitors (i.e., vanadates, molybdates, tungstates, phosphates, borates) and cathodic inhibitors (i.e., Ce, Y, La, Eu, Gd and Nd) those are proved to be more suitable to substitute the toxic inhibitor of chromates (Tayler and Chambers, 2009). The inhibition efficiency of sodium metavanadate for controlling corrosion of carbon steel in an aqueous solution containing chloride ion was evaluated (Sribharathy & Rajendran, 2012). However, a few studies are reported about the corrosion behavior of different steels used in Nepal in aqueous solutions (Bhattarai, 2008, 2009; Bhattarai *et al.,* 2007) and corrosion of buried corrosive nature of soils towards the galvanized-steel as well as cast-iron pipes used for the drinking water supply in Nepal were also reported by some researchers (Bhandari *et al.,* 2013; Bhattarai *et al.,* 2016; Bhattarai, 2013; Dahal *et al.,* 2014; Dhakal *et al.,* 2014; Gautam and Bhattarai, 2013). In this context, present research work was focused to study the effect of sodium

tungstate as a green corrosion inhibitor on the corrosion behavior of the mild steel used Nepal in different aggressive aqueous media.

The main objective of the present research work is to study the effect of sodium tungstate on the corrosion behavior of mild steel grille sheet which is used as the materials for the window grille production, in 0.5 M NaCl solution open to air at 25±1°C. An attempt was also made to study the effects of varying concentrations of sodium tungstate on the passivation behavior of the mild steel grille sheet in 0.5 M NaCl solution at 25±1°C employing corrosion tests, inhibition mechanism and electrochemical measurements.

Materials and Methods

Corrosion tests of the steel grille sheet specimens were carried out in 0.5 M NaCl solution open to air at 25±1°C containing different concentrations of 100, 200, 400 and 800 ppm of sodium tungstate. It was also estimated the corrosion rate of the mild steel grille sheet in 1 M HCl and 1 M NaOH solutions for comparision. Prior to corrosion test, the surface of each grille sheet specimen was mechanically polished with silicon carbide paper grit numbers 200-1500, rinsed with acetone and dried by air blowing in order to obtain reproducible results. Weight loss method was used to estimate the corrosion rate of the test specimens. The corrosion rate measurement by weight loss method was done two times or more so as to obtained average corrosion rate. The average corrosion rate of the individual grille sheet specimen was estimated using following equation 1 (Bhattarai, 2010).

$$\text{Corrosion Rate (mm.y}^{-1}) = \frac{\Delta w \times 8760 \times 10}{d \times A \times t} \quad (1)$$

Where, Δw is the weight loss of the grille sheet specimen in gram, d is the density of the tested specimen in g/cm^3, A is area of the sample specimen in cm^2 and t is time of immersion in hour.

Inhibitors efficiencies based on the corrosion rate ($IE_{CR,\%}$) and the degree of surface coverage of the inhibitor molecule (θ) were also calculated using following equations 2 and 3, respectively (Hegazy *et al.,* 2012).

$$IE_{CR,\%} = \frac{CR_{(o)} - CR_{(inhib.)}}{CR_{(o)}} \times 100 \quad (2)$$

$$\theta = \frac{CR_{(o)} - CR_{(inhib.)}}{CR_{(o)}} \quad (3)$$

Corrosion inhibition mechanism was also studied using Langmuir adsorption model. The Langmuir adsorption isotherm equation (Satapathy *et al.,* 2009) used here is expressed in equation 4 where, $C_{inhib.}$ is the inhibitor concentration and K_{ads} is the adsorptive equilibrium constant. The equation (4) was used to study the adsorption

mechanism of the green inhibitor of sodium tungstate for the passivity of the grill steel sheet in 0.5 M NaCl solution 25±1°C.

$$\frac{C_{inh.}}{\theta} = \left(\frac{1}{K_{ads}}\right) + C_{inh.} \qquad (4)$$

Where, $CR_{(o)}$ and $CR_{(inhib.)}$ are the corrosion rates in the absence and presence of inhibitor, respectively. $C_{inh.}$ is the inhibitor concentration, θ is the surface coverage and K_{ads} is the adsorptive equilibrium constant.

The K_{ads} value was estimated from the intercept of a straight line obtained by plotting $C_{inhib.}/\theta$ vs $C_{inhib.}$. The K_{ads} value was used to estimate the standard free energy of adsorption (ΔG°_{ads}) using equations 5 and 6 (Cases and Villieras, 1992).

$$K_{ads} = \frac{1}{55.5} \exp\left(-\frac{\Delta G^{o}_{ads}}{RT}\right) \qquad (5)$$

$$\Delta G^{o}_{ads} = -RT \ln\left(55.5 \times K_{ads}\right) \qquad (6)$$

Where, R is gas constant, T is temperature and the value of 55.5 is the molar concentration of water in solution.

The test specimen was immersed in electrolytes in such a way that about 72 mm^2 area of the specimen was used for electrochemical measurements using O-ring for anodic potentiodynamic polarization measurement in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air containing different concentrations of 100, 200, 400 & 800 ppm sodium tungstate at 25±1°C. Open circuit potential (OCP) of the mild steel specimens were measured using a simple potentiometer (Osaw digital potentiometer) in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C containing different concentrations of sodium tungstate. The readings were taken after immersion of the sample specimen in the electrolytic solutions for 72 hours. Potentiodynamic anodic polarization curves for the grille sheet of the mild steel were obtained in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C using a potentiostat/galvanostat (Hokuto Denki HA-151 Model). The potentiodynamic anodic polarization measurement was carry out for the grille sheet specimens after immersion for 30 minutes when the open circuit potential of the specimens became almost steady. A saturated calomel electrode (SCE), a platinum mesh and grille sheet were used as the reference, counter and working electrodes, respectively.

Results and Discussion

Corrosion rate

The corrosion rate of the mild steel sheet, which is generally used for the production of grille in Nepal, was estimated after immersion for 216 hours in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C using weight loss method. The corrosion rate for each sample specimen

was estimated two times or more and average corrosion rate was calculated which is shown in Fig. 1. The average corrosion rates of the mild steel sheet were about 1.27×10^1 mm/y, 4.21×10^{-2} mm/y and 3.50×10^{-3} mm/y in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions, respectively, as tabulated in Table 1 also. The corrosion rate of the grille sheet in 1 M HCl solution was nearly three orders of magnitude higher than its corrosion rate in 0.5 M NaCl solution and four orders of magnitude higher than its corrosion rate in 1 M NaOH solution. These results revealed that the corrosion rate of the mild steel sheet of Nepal in acidic 1 M HCl solution is significantly higher than those in the near neutral 0.5 M NaCl and strong alkaline 1 M NaOH solutions open to air at room temperature. This is mostly due to the pH effects of the aqueous solution, because iron or steel is generally passive in the aqueous solution having pH value in the range of 9.6 to 13.5 (Bhattarai, 2010).
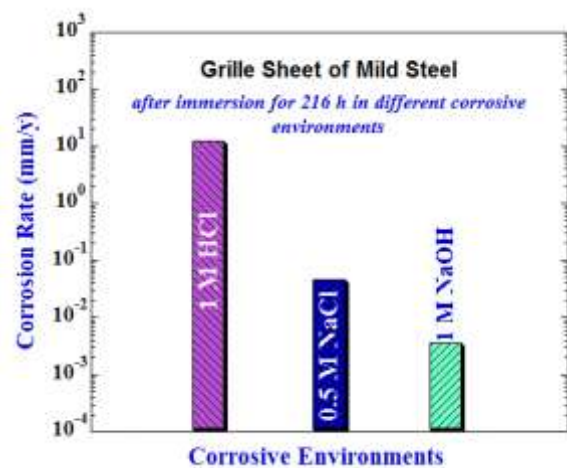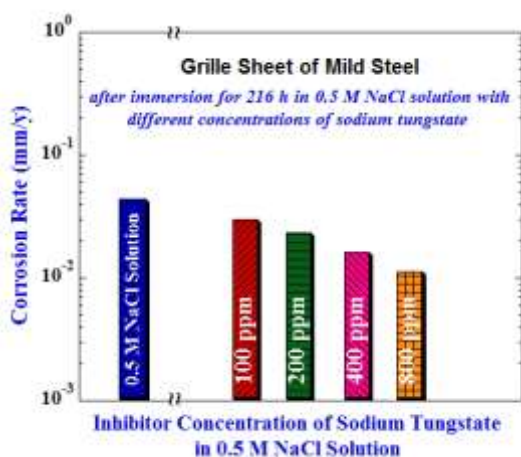

Fig. 1: Corrosion rate of the mild steel sheet in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C

Effect of sodium tungstate as a green corrosion inhibitor

The effect of 100, 200, 400 and 800 ppm sodium tungstate for the steel grille sheet was studied in 0.5 M NaCl solution open to air at 25±1°C. The corrosion rate of the mild steel sheet is decreased with increasing the concentrations of sodium tungstate as depicted in Fig. 2 and also summarized in Table 1. The result revealed that sodium tungstate is an effective corrosion inhibitor to increase the corrosion resistance properties of the steel grille sheet in 0.5 M NaCl solution.

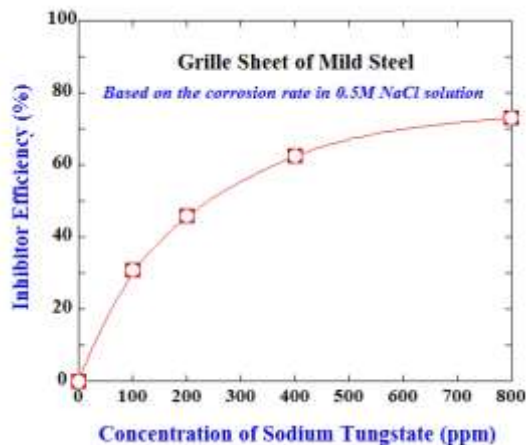### Inhibitor efficiency based on the corrosion rate

The inhibition efficiency of different concentrations of sodium tungstate was studied in 0.5 M NaCl solution and the results are shown in Fig. 3 and tabulated in Table 1. Initially the inhibition efficiency increased steeply with increasing the concentration of sodium tungstate up to 400 ppm and the maximum inhibition efficiency of 73% was obtained at 800 ppm. This behavior may be attributed that the use of sodium tungstate is acting as adsorption inhibitor.

Fig. 2: Changes in the corrosion rates of the mild steel sheet in 0.5 M NaCl solution open to air at 25±1°C, as a function of the concentration of sodium tungstate

*Mechanism of corrosion inhibition*

It is reasonable to consider quasi-equilibrium adsorption in thermodynamic using an appropriate adsorption isotherm to explain the mechanism of corrosion inhibition. The process of inhibitor adsorption on the surface of mild steel sheet can be described by different isotherms, from which Langmuir is the simplest and is based on the assumption that all adsorption sites are equivalent and the particle binding occurs independently from nearby sites being occupied or not (Mortimer, 2008).

Fig. 3. Changes in the inhibitor efficiency based on the corrosion rate of the mild steel sheet in 0.5 M NaCl solution open to air at 25±1°C, as a function of the concentration of sodium tungstate

Fig. 4 shows the relationship between the ratio of inhibitor concentration to surface coverage ($C_{inh.}/\theta$) and inhibitor concentration ($C_{inh.}$) for the corrosion inhibitor of sodium tungstate in 0.5 M NaCl solution open to air at 25±1°C. The linear correlation coefficient (R^2) was used to choose the isotherm that best fit experimental data, because it was found almost equal to unity (i.e., $R^2 = 0.9987$) as shown in Fig. 4. The result indicated that the adsorption process obeyed Langmuir adsorption isotherm to explain the

corrosion inhibition mechanism on the surface of the mild steel sheet by green inhibitor of sodium tungstate.
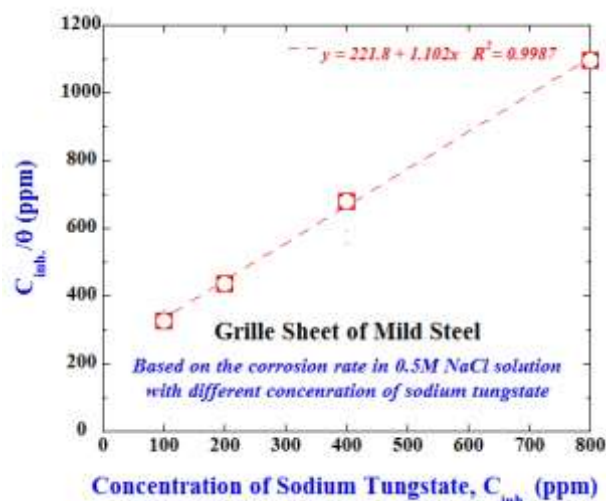
Fig. 4. Langmuir's adsorption plot for the mild steel sheet in 0.5 M NaCl solution open to air at 25±1°C in presence of different concentrations of sodium tungstate

Electrochemical measurements

Electrochemical measurements (i.e. open circuit potential and anodic polarization) were carried out for a better understanding of the corrosion behavior as well as the anodic passivity of Nepalese mild steel sheet in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C in absence and presence of sodium tungstate as a green corrosion inhibitor. A grille sheet specimen, saturated calomel electrode (SCE) and platinum mesh were used as working, reference and counter electrodes, respectively. All the potentials given in this paper are relative to SCE.

Open circuit potential

Figure 5 shows the changes in open circuit potential (OCP) for the mild steel sheet after immersion for 72 hours in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C, as a function of immersion time. The change in the open circuit potentials for the examined steel sheet is not significant with immersion time in 1 M HCl solution although it is slightly shifted to positive (less noble) direction after immersion for 72 h as shown in Fig. 5. On the other hand, the OCP is shifted slightly to more negative (less noble) direction with immersion time in 0.5 M NaCl solution although the OCP is remarkably shifted to more positive (more noble) direction in 1 M NaOH solution as shown in Fig. 5. Consequently, the OCP of the grille sheet made by mild steel was recorded more negative value (i.e. about -659 mV vs SCE) in 0.5 M NaCl solution than that in 1 M HCl solution (i. e. –496 mV vs SCE) after immersion for 72 hours. However, in 1 M NaOH solution, the OCP is more positive value (i.e. about –176 mV vs SCE) than that in 0.5 M NaCl solution as tabulated in Table 1. These results revealed that the grille sheet used to carry out present research work is found to be more passive in 1 M NaOH solution than in 1 M HCl and 0.5 M NaCl solutions. This is

mostly due to the formation of passive films on the surface of steels/iron at pH range of 9.6-13.6 (Bhattarai, 2010; Revie & Uhlig, 2008).
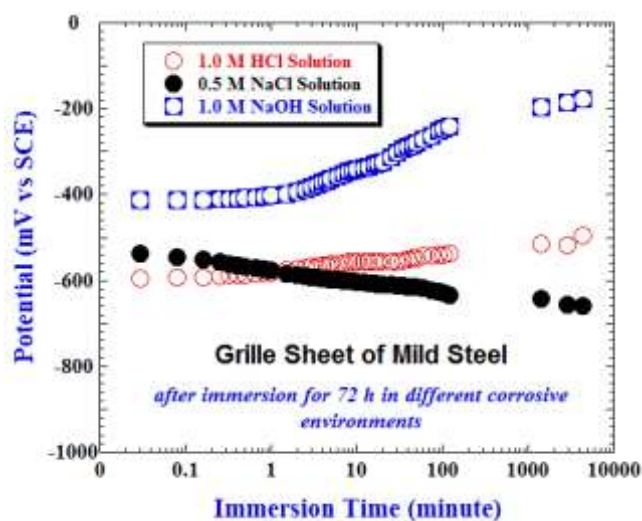

Fig. 5. Changes of the OCP for the mild steel sheet in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C, as a function of immersion time

The effect of different concentrations of sodium tungstate inhibitor was studied after immersion up to 72 hours in 0.5 M NaCl solution open to air at 25±1°C to study the corrosion behavior and the passivity of the grille sheet made by mild steel. The open circuit potentials for the mild steel sheet after immersion up to 72 h in 0.5 M NaCl solution, open to air at 25±1°C in the absence and the presence of different concentrations of sodium tungstate are shown in Fig. 6 and summarized in Table 1 also. It is clearly shows that the OCP of the steel sheet after immersion for 72 h in 0.5 M NaCl solution with different concentrations of sodium tungstate corrosion inhibitor is shifted to more positive direction with increasing the concentrations of the inhibitor.
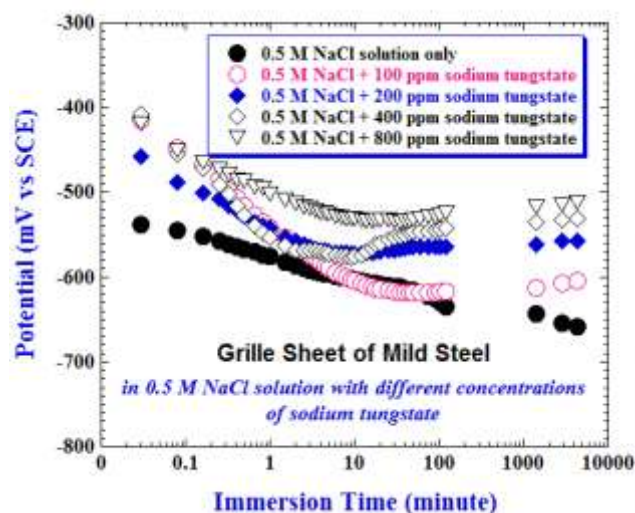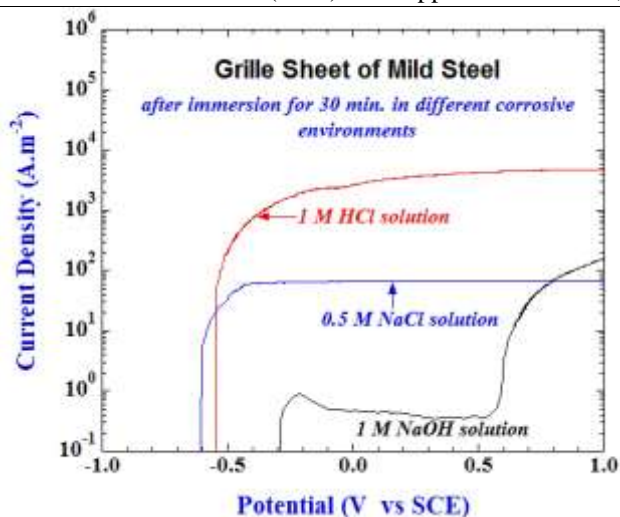

Fig. 6: The effects of different concentrations of the corrosion inhibitor of sodium tungstate in open circuit potential for the mild steel sheet in 0.5 M NaCl solution open to air at 25±1°C, as a function of immersion time

Anodic polarization curve

In order to study the passivation of the grille sheet, the potentiodynamic anodic polarization measurement was carried out in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C. Figure 7 shows the potentiodynamic anodic polarization curves for the grille steel specimens in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C. Spontaneous passivation occurs for the grille sheet specimens in a wide potential region up to 1000 mV (SCE) after potentiodynamic polarization in 1 M HCl and 0.5 M NaCl solutions while the grille sheet in alkaline 1 M NaOH solution shows the active-passive transition and transpassive dissolution. In 1 M NaOH solution, the grill sheet is active in the potential range of –290 to –210 mV (SCE) while it is passive at the potentials between –210 to 550 mV (SCE). Transpassivity was clearly seen after polarization at about 550 mV (SCE) or more anodic polarization potential probably due to the formation of soluble ferrate (FeO_4^{2-}) in 1 M NaOH solution as shown in Fig. 7.

Table 1: Corrosion rate (CR) of the mild steel sheet in different corrosive environments in presence and absence of sodium tungstate as a green corrosion inhibitor

S. N.	Inhibitor concentration (ppm)	CR in 1M HCl soln. (mm/y)	CR in 1 M NaOH soln. (mm/y)	CR in 0.5 M NaCl soln. (mm/y)	CR in 0.5 M NaCl + tungstate (mm/y)	Inhibitor efficiency (%)	Corrosion potential (mV vs SCE)
1	-	12.7230	0.0035	0.0421			–659 (0.5M NaCl)
2	100				0.0292	30.71	–605
3	200				0.0228	45.75	–557
4	400				0.0158	62.41	–531
5	800				0.0114	73.00	–512

Fig.7: Anodic polarization curves measured for the grille sheet of Nepal after immersion for 30 minutes in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 25±1°C

The anodic passive current densities of the grill sheet specimens were in the ranges of $2\text{-}5 \times 10^1$ A.m^{-2}, $6\text{-}7 \times 10^{-1}$ A.m^{-2} and $4\text{-}6 \times 10^{-3}$ A.m^{-2} in 1 M HCl, 0.5 M NaCl and 1 M NaOH solutions, respectively. It is found that the anodic passive current density of the grille sheet in alkaline 1 M NaOH solution is significantly lower than those in 1 M HCl and 0.5 M NaCl solutions. The anodic passive current density in 1 M NaOH solution is nearly two orders of magnitude lower than in 0.5 M NaCl solution and is nearly four orders of magnitude lower than in 1 M HCl solution. Consequently, it can be said that the more stable anodic passive film is formed on the surface of the grille sheet after anodic polarization in alkaline 1 M NaOH solution than those in strong acidic 1 M HCl as well as the near neutral 0.5 M NaCl solutions and hence the corrosion resistance property of the grille sheet specimen in 1 M NaOH is higher than those in 1 M HCl and 0.5 M NaCl as shown in Fig. 1 also.

Conclusions

The corrosion inhibition effect of sodium tungstate on the passivation behavior of the grille sheet used in Nepal made by the mild steel in different corrosive media open to air at 25±1°C was carried out using corrosion tests and electrochemical measurements. From the results and discussion following conclusions are drawn.

The corrosion rate of the mild steel sheet in 1 M HCl solution was nearly three and four orders of magnitude higher than the corrosion rates in 0.5 M NaCl and 1 M NaOH solutions, respectively.

The corrosion rate of the steel grille sheet was decreased with increasing the concentrations of sodium tungstate up to the concentration of 800 ppm in 0.5 M NaCl solution open to air at 25±1°C.

The inhibition efficiency was increased with increasing the inhibitors concentration which indicated that the used of

three inhibitors are acting as adsorption inhibitors in 0.5 M NaCl solution.

The corrosion inhibition efficiency was increased with increasing the concentration of sodium tungstate in 0.5 M NaCl solution and follows the Langmuir adsorption model.

The open circuit potential of the steel grille sheet was more negative value in 0.5 M NaCl solution than that in 1 M HCl solution, whereas more positive potential value was observed in 1 M NaOH than in 0.5 M NaCl after immersion for 2 h.

The open circuit potential of the steel grille sheet in presence of sodium tungstate inhibitor was shifted to more positive direction with increasing the concentrations of the inhibitor in 0.5 M NaCl solution.

The mild steel sheet was passivated spontaneously in a wide range of the anodic potentials in 1 M HCl and 0.5 M NaCl solutions, whereas it showed the active-passive transition and transpassive dissolution in 1 M NaOH solution. The anodic passive current density in 1 M NaOH solution is nearly two orders of magnitude lower than in 0.5 M NaCl solution and is nearly four orders of magnitude lower than in 1 M HCl solution.

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