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Research Article

Study the Corrosion Inhibition Effect of Sodium Tungstate for Chromium-based Ternary Alloys in 0.5 M NaCl solution

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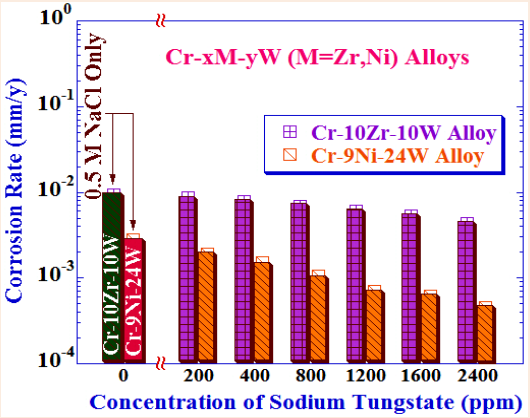
Abstract

Sodium tungstate inhibitor enhanced the corrosion resistance properties of the sputter-deposited chromium-based, Cr-xM-yW (M=Zr, Ni) alloys in 0.5 M NaCl solution, open to air at 25±1ºC. Corrosion inhibition mechanism of the alloys by sodium tungstate was explained using Langmuir isotherm. Standard free energy of sodium tungstate inhibitor adsorption on the surface of Cr-10Zr-10W and Cr-9Ni-24W alloys was found to be -22.55 KJ/mol and -26.31 KJ/mol, respectively which indicated that sodium tungstate is strongly adsorbed on the alloy surfaces by physical adsorption, not by the chemisorption. Hence the corrosion resistance properties of the Cr-xM-yW (M=Zr, Ni) alloys was increased with increasing sodium tungstate inhibitor concentrations. From the corrosion potential measurements, it is concluded that sodium tungstate acts as an anodic corrosion inhibitor, because the corrosion potential of the alloys was shifted to more positive direction with increasing the concentrations of the inhibitor in 0.5 M NaCl.



Keywords: Tungstate inhibitor, corrosion mechanism, corrosion potential, sputtered chromium-based alloy, 0.5 M NaCl

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Introduction

Corrosion control method becoming a subject of tremendous technological and industrial significances in these times. It is a fundamental academic and research concerns of corrosion scientists, engineers and technologists to control the corrosion of metals and alloys using environmental friendly corrosion inhibitors [1, 2]. The corrosion problems can be solved or minimized using different methods. Nowadays, the use of eco-friendly green corrosion inhibitors in corrosive environments is becoming one of the widely practiced corrosion controlled methods. The corrosion inhibitor is a chemical substance that when added in a small amount to a corrosive environment, effectively decreases the corrosion rate of the metallic materials exposed to that environment [1]. The corrosion inhibitors interferes the corrosion kinetics and thereby control the corrosion rate of the metals and alloys.

Corrosion control of metals and alloys is an important activity of technical, economical and environmental importance. The use of inhibitors is one of the best options of protecting metals and alloys against corrosion. The toxicity of organic and inorganic corrosion inhibitors to the environment has motivated the search for safer corrosion inhibitors such as green corrosion inhibitors as other more environmental friendly corrosion inhibitors. Most of the environmental friendly green inhibitors are biodegradable and do not contain toxic metals, or other compounds. Investigations of corrosion-inhibiting abilities of inorganic radicals or ions, e.g., nitrites, molybdates, tungstates, polyphosphates, nitrates etc have shown that these radicals or ions are inexpensive, readily available and renewable sources of materials in addition to being environmental friendly and ecologically acceptable. Need for more effective inhibitors have incited corrosion scientists, engineers and technologists to develop new corrosion resistance materials used in aggressive environments.

Several researches have indicated that some inorganic compounds can be used as environmental friendly green corrosion inhibitor, because they form complexes with metal ions on metal or alloy surface. The complex occupies a large surface area, thereby blanketing the surface and protecting the metals or alloys from corrosive ions of the environment. The corrosion inhibition of various metals and alloys by a lot of chemical compounds has been investigated by various researchers in the past [3-14]. They found that corrosion inhibitors having their specified radicals and ions were effective to control the corrosion. However, all of these corrosion inhibitors are not eco-friendly green corrosion inhibitors. For example, chromate (VI) was reported as one of the most effective inhibitors, but it is toxic to human beings [14, 15]. Therefore, nowadays chromium (VI) is not generally practiced as a green corrosion inhibitor. There is great interest in replacing it by effective and non-hazardous alternatives eco-friendly green corrosion inhibitors and hence many alternative eco-friendly green organic [8-12, 16], inorganic [16-18] and corrosion inhibitors are developed. Tungstates, molybdates, nitrites are now being increasingly used as environmental friendly green inorganic corrosion inhibitor due to their low order of toxicity [19-24].

The green corrosion inhibitors may act in a number of ways: they may restrict the rate of anodic or/and cathodic reactions by simply blocking active sites on the metal and alloy surfaces. Alternatively, it may act by increasing the potential of the metal and alloy surface so that the metal and alloy enters the passivation region where passive film forms. A further mode of action of some inhibitors is that the inhibiting compound contributes to the formation of a thin layer on the surface which repressed the corrosion process. The main strategy of the green corrosion inhibitors use is to isolate effectively the metal and alloy from corrosive environments without their adverse effect to our societal health as well as environment in the long-time [25]. Corrosion inhibition mechanism occurs via adsorption of their molecules on the corroding metal and alloy surfaces and hence the inhibition efficiency depends on the mechanical, structural, chemical and thermodynamic characteristics of the adsorption layers formed under particular condition. Corrosionists' activities in recent times are pitched towards finding alternative environmental friendly green corrosion inhibitors to replace the toxic and hazardous radicals of compound like chromates.

It is meaningful for mentioning here that the sputter-deposited amorphous /nanocrystalline W-xZr-yCr [26-32] and W-xCr-yNi [26, 33-36] alloys were spontaneously passivated showing higher corrosion resistance properties than those of their alloy-constituting elements in different aggressive acidic to alkaline solutions. In this context, the present work is focused to study the effects of the environmental friendly green corrosion inhibitor of sodium tungsten on the corrosion behavior and inhibition mechanism of two sputter-deposited chromium-based ternary Cr-10Zr-10W and Cr-9Ni-24W alloys in 0.5 M NaCl solution, open to air at 25±1°C using corrosion tests, inhibition mechanism and open circuit (or corrosion) potential measurements.

Experimental

Cr-xM-yW (M=Zr, Ni) alloys

The sputter-deposited ternary chromium-based Cr-xM-yW (M=Zr, Ni) alloys were used in the present research work. The instrument used and conditions subjected for the sputter deposition of Cr-xM-yW (M=Zr, Ni) alloys were same as those described elsewhere [27, 31, 33, 36]. X–ray diffractometer with CuK_α radiation at θ-2θ mode and electron probe microanalysis were used to determine the structure and composition of the alloys, respectively. Apparent grain size of the alloys was estimated using Scherrer's formula [37] from the most intense XRD peak.

Corrosion rate measurement

The corrosion test of the Cr-xM-yW (M=Zr, Ni) alloys was carried out after immersion for 240 h in 0.5 M NaCl solution open to air at 25±1°C with different concentrations (i.e. 200, 400, 800, 1200, 1600 and 2400 ppm) of sodium tungstate. Before each corrosion test, the alloy specimens were polished mechanically using silicon carbide paper having grit number 1500 in cyclohexane. The polished alloy specimens were rinsed with acetone and dried in air to obtain reproducible result. The weight loss method as described elsewhere [2] was used to estimate the average corrosion rate of the alloy specimens using equation (1). The average corrosion rate measurement was carried out two times or more so as to obtained delimited results.

$$\text{Corrosion Rate (mm/y)} = \frac{\Delta w \times 8760 \times 10}{d \times A \times t} \tag{1}$$

Where, Δw is the weight loss of the alloy specimen in gram (g), d is the density of the alloy specimen in g/cm^3, A is area of the alloy specimen in cm^2 and t is the time of immersion in hour.

Corrosion inhibition efficiency and mechanism studies

In order to explain the corrosion inhibition mechanism of the Cr-xM-yW (M=Zr, Ni) alloys in 0.5 M NaCl solution in presence of sodium tungstate, the percentage inhibition efficiency (% IE) and the degree of surface coverage, θ [38] were estimated using equations (2) and (3), respectively, where, $CR_{(unhib.)}$ and $CR_{(inhib.)}$ are the corrosion rates of the alloys in absence and presence of sodium tungstate, respectively. The Langmuir adsorption isotherm as given in equation (4) was used to study the corrosion inhibition mechanism. The adsorptive equilibrium constant (K_{ads}) value was estimated from the intercept of a straight line obtained by plotting $C_{inhib.} / \theta$ vs $C_{inhib.}$ Furthermore, the K_{ads} value was used to estimate the standard free energy change of adsorption (ΔG^{o}_{ads}) using equations (5) and (6) [38].

$$\%\,IE = \frac{CR_{(unhib)} - CR_{(inhib.)}}{CR_{(unhibit)}} \times 100 \tag{2}$$

$$\theta = \frac{CR_{(unhib.)} - CR_{(inhib.)}}{CR_{(unhib.)}} \tag{3}$$

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

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

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

Where, $C_{inhib.}$ is the inhibitor concentration (g/mol), R is gas constant (J/K.mol), T is absolute temperature (K) and the value of 55.5 is the molar concentration of water in solution [5].

Corrosion potential measurement

The corrosion potential of the sputtered Cr-xM-yW (M=Zr, Ni) alloys was measured in 0.5 M NaCl solution open to air at 25±1^{o}C containing different concentrations (i.e. 200, 400, 800, 1200, 1600 and 2400 ppm) of sodium tungstate as a green corrosion inhibitor. The alloy specimens were first mechanically polished by silicon carbide paper having grit number 1500 in cyclohexane, rinsed with acetone and dried in air to obtain reproducible results. Then, the test alloy specimen was clipped by a sample holder that was made by welding the crocodile pin with a titanium rod. The readings were taken immediately after immersion of the alloy specimen in the electrolytic solution until 72 hours at different time intervals. The corrosion potential measurement was repeated two times or more. A saturated calomel electrode (SCE) and the sputter-deposited alloy specimens were used as reference and working electrodes, respectively.

Results and Discussion

Characterization of alloys

The composition, apparent grain size, structure and corrosion rate of the sputter-deposited Cr-xM-yW (M=Zr, Ni) alloys including alloy-constituting elements are summarized in **Table 1** [39-44]. Alloy composition hereafter is denoted by atomic percentage (at. %).

Table 1 Composition, apparent grain size, structure and corrosion rate of the sputter-deposited Cr-xM-yW (M=Zr, Ni) alloys including alloy-constituting elements.

Alloys & Alloying Elements	Cr Content (at.%)	Zr Content (at.%)	Ni Content (at.%)	W Content (at.%)	Apparent Grain Size (nm)	Structure*	Corrosion Rate in 0.5 M NaCl, 25°C (mm/y)
Cr	100	-	-	-	18 [39]	Nano-cryst.	0.0074 [40]
W	-	-	-	100	18 [41]	Nano-cryst.	0.0250 [42]
Cr-10Zr-10W	79.98	9.94	-	10.08	29	Nano-cryst.	0.0095
Cr-9Ni-24W	66.93	-	9.08	23.99	1.5	Amor.	0.0028
Zr	-	100	-	-	23 [41]	Nano-cryst.	0.0031 [43]
Ni	-	-	100	-	19 [44]	Nano-cryst.	0.3021 [44]

**Nanocryst. = Nanocrystalline; Amor. = Amorphous*

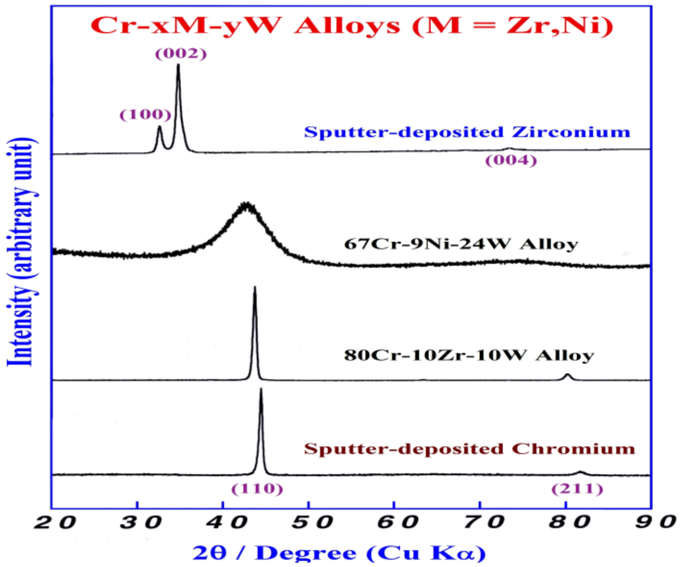

Figure 1 XRD patterns of the sputter-deposited chromium-based ternary alloys and their alloy-constituting elements

Inhibition effects of sodium tungstate in corrosion rate

The effect of different concentrations (i.e., 200, 400, 800, 1200, 1600 and 2400 ppm) of sodium tungstate in the corrosion rate of the sputter-deposited Cr-xM-yW (M=Zr, Ni) alloys was estimated after immersion for 240 h in 0.5 M NaCl solution open to air at 25±1ºC. The corrosion rate of both Cr-10Zr-10W and Cr-9Ni-24W alloys was decreased with increasing concentrations of sodium tungstate between 200 to 2400 ppm in 0.5 M NaCl solution as depicted in **Figure 2**. The corrosion rate of the alloys was found to be lowest in 2400 ppm of sodium tungstate in 0.5 M NaCl solution and it is nearly one order of magnitude lower than in 0.5 M NaCl solution in addition of 2400 ppm sodium tungstate for the Cr-9Ni-24W alloy particularly. However, the corrosion rate of the Cr-10Zr-10W alloy is not significantly decreased as for Cr-9Ni-24W alloy and hence the corrosion resistance property of the Cr-10Zr-10W alloy is lower than that for the Cr-9Ni-24W alloy in absence and presence of sodium tungstate in 0.5 M NaCl. These results revealed that the use of sodium tungstate as an environmental friendly green corrosion inhibitor enhanced to increase the corrosion resistance properties of the sputter-deposited ternary Cr-10Zr-10W and Cr-9Ni-24W alloys in near neutral 0.5 M NaCl solution, open to air at 25±1ºC.

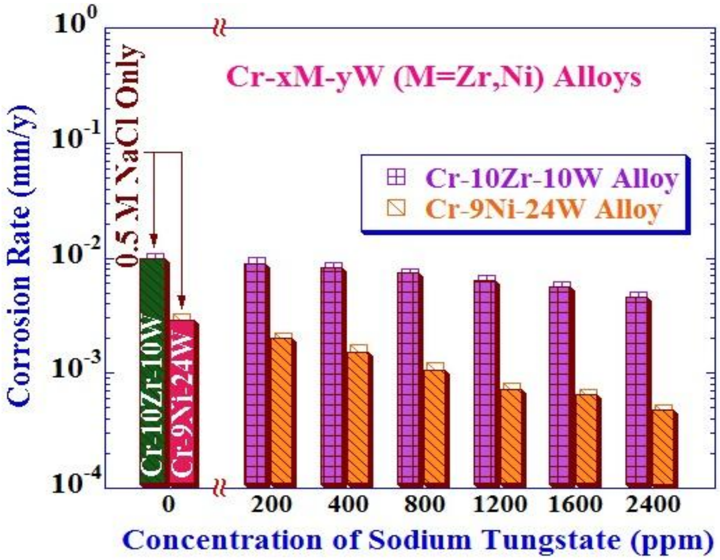

Figure 2 Changes in the corrosion rates of the sputter-deposited chromium-based ternary alloys after immersion for 240 hours in 0.5 M NaCl solution, open to air at 25±1°C, as a function of sodium tungstate concentration

Corrosion inhibition efficiency and mechanism

The inhibition efficiency of sodium tungstate in 0.5 M NaCl solution for the sputter-deposited Cr-xM-yW (M=Zr, Ni) alloys is increased with increasing sodium tungstate concentrations as shown in **Figure 3 (a)**. The maximum inhibition efficiency of about 75-84 % was reached between 1200 to 2400 ppm of sodium tungstate in 0.5 M NaCl solution for the Cr-9Ni-24W alloy whereas it was about 37-55 % for the Cr-10Zr-10W alloy as shown in **Figure 3 (a)**. It was found that the most efficient corrosion inhibitor concentration of sodium tungstate was found to be at 2400 ppm for both the alloys in 0.5 M NaCl solution. The inhibition action of sodium tungstate was found to obey the Langmuir isotherm model of adsorption. Consequently, the increase in the percentage corrosion inhibition efficiency with inhibitor concentrations indicated that the sodium tungstate acting as adsorption inhibitor to decrease the corrosion rate of the sputter-deposited Cr-10Zr-10W and Cr-9Ni-24W alloys in 0.5 M NaCl solution, open to air at 25±1ºC.

The Langmuir adsorption isotherm model was performed to have more insights into the corrosion inhibition mechanism by sodium tungstate on the sputter-deposited Cr-10Zr-10W and Cr-9Ni-24W alloys in 0.5 M NaCl solution. In order to obtain adsorption isotherm to explain the corrosion inhibition mechanism of sodium tungstate for the Cr-10Zr-10W and Cr-9Ni-24W alloys, the θ value was estimated from the weight loss measurement at different concentrations of sodium tungstate**. Figure 3 (b)** shows the relationship between C_{inhib} / θ and concentration of sodium tungstate ($C_{inhib.}$) for the sputter-deposited Cr-xM-yW (M=Zr, Ni) alloys in 0.5 M NaCl solution. The linear correlation coefficient (R^2) and the slope of the straight line for both the alloys were found to almost unity. These results indicated that the adsorption process obeyed Langmuir adsorption isotherm to explain the corrosion inhibition mechanism on the surface of the alloys by sodium tungstate in 0.5 M NaCl solution, open to air at 25±1°C.

It is meaningful for mentioning here that there is no interaction between the adsorbed corrosion inhibitor molecules, the energy of adsorption is independent with θ, the alloy surfaces contain a fixed number of adsorption sites and each site holds one adsorbed species according to the Langmuir isotherm model [21]. In literature, it was reported that the high K_{ads} value (i.e., >100 mol/g) attributes to stronger and more stable adsorbed layer formation on the metals or/and alloys surfaces [45]. It was also reported that the ΔG^{o}_{ads} values of metals or alloys around -20 KJ/mol or lower indicate adsorption of inhibitors onto material surface with electrostatic interaction is due to physical adsorption, while those around or higher (more negative) than -40 KJ/mol involve charge sharing between inhibitor molecules and material surfaces is of chemisorption [46]. Negative values of ΔG^{o}_{ads} ensure the spontaneity of the adsorption process and stability of the adsorbed layer of the corrosion inhibitors on the materials surfaces.

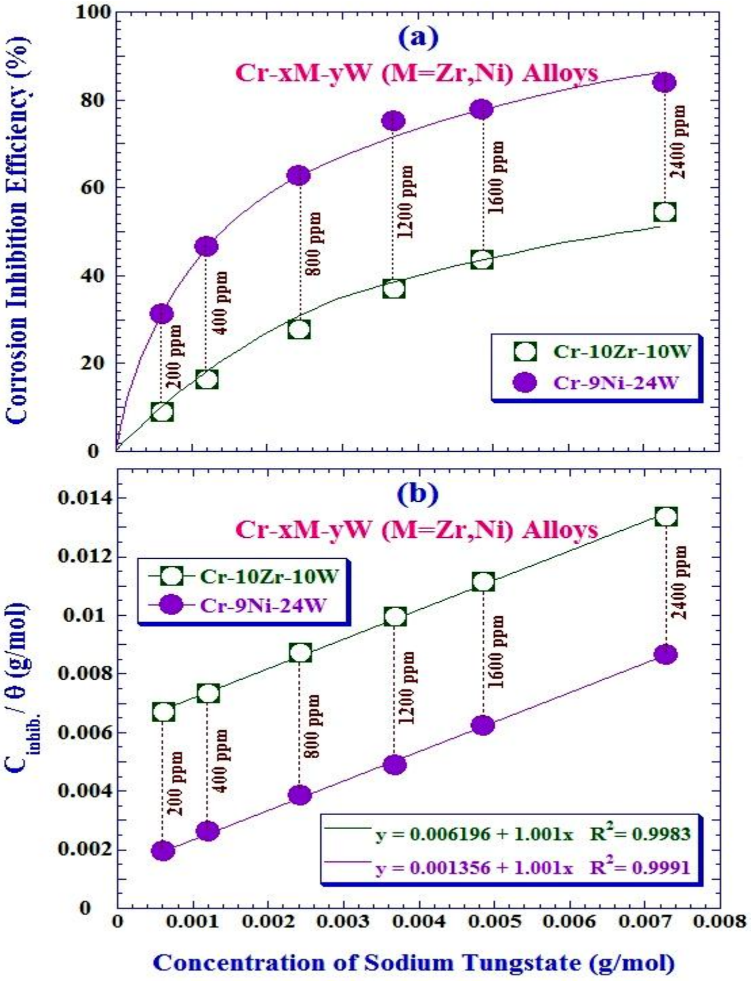

Figure 3 (a) Changes in the corrosion inhibition efficiency and (b) Langmuir plot for the sputter-deposited chromium-based ternary alloys after immersion for 240 hours in 0.5 M NaCl solution, open to air at 25±1°C, as a function of sodium tungstate concentration

In the present study, the ΔG^{o}_{ads} values for sodium tungstate as a corrosion inhibitor in 0.5 M NaCl solution, open to air at 25±1ºC for the sputter-deposited Cr-10Zr-10W and Cr-9Ni-24W alloys were found to be -22.55 and -26.31 KJ/mol , respectively, that are consistent with literatures for physical type of adsorption. Hence authenticates physical adsorption of sodium tungstate on the surface of Cr-10Zr-10W and Cr-9Ni-24W alloys. This implies that the corrosion inhibitor of sodium tungstate adhere on the surfaces of the Cr-xM-yW (M = Zr, Ni) alloys and so give more efficient inhibition effects to decrease the corrosion rate of the ternary alloys in 0.5 M NaCl solution, open to air at 25±1ºC.

Corrosion potential

The corrosion potential ($\phi_{corr.}$) or open circuit potential (OCP) measurement was carried out for a better understanding of the corrosion behavior of the Cr-xM-yW (M=Zr, Ni) alloys in 0.5 M NaCl solution, open to air at 25±1ºC in absence and presence of sodium tungstate. **Figures 4 (a)** and **4 (b)** show the changes in the open circuit potential for the Cr-10Zr-10W and Cr-9Ni-24W alloy specimens, respectively, after immersion for 72 hours in 0.5 M NaCl solution at 25ºC in absence and presence of sodium tungstate concentrations.

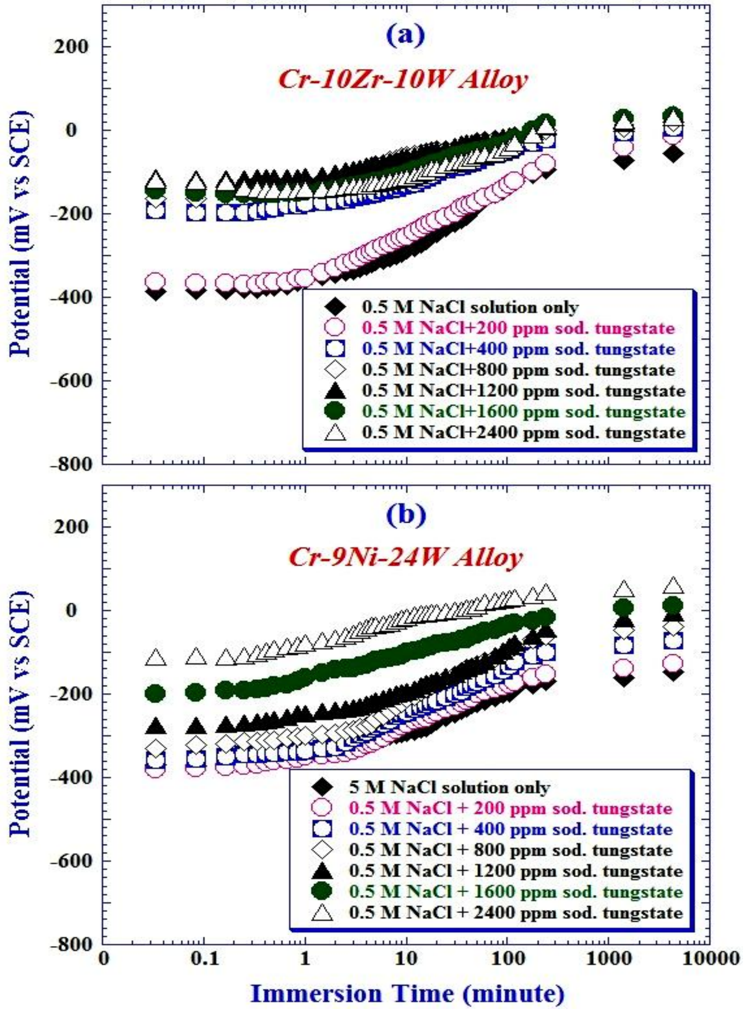

Figure 4 Changes in open circuit potential of the sputter-deposited (a) Cr-10Zr-10W and (b) Cr-9Ni-24W alloys after immersion for 72 hours in 0.5 M NaCl solution, open to air at 25±1°C in absence and presence of sodium tungstate as corrosion inhibitor

It is clearly showed that the corrosion potential of the alloys in 0.5 M NaCl solution with the addition of sodium nitrite concentrations is shifted to more positive or noble direction with increasing the concentrations of sodium tungstate after immersion for 72 hours as shown in **Figure 4**. In particular, the shifting of the corrosion potential towards the more positive direction with increasing the concentration of the sodium tungstate is more clearly observed for the Cr-9Ni-24W alloy than for the Cr-10Zr-10W alloy which is consistent with the corrosion rate result as shown above in **Figure 2**. These results revealed that the environmental friendly green corrosion inhibitor of sodium tungstate acts as the anodic inhibitor for the ternary Cr-xM-yW (M=Zr, Ni) alloys in near neutral 0.5 M NaCl solution.

Conclusions

The effect of sodium tungstate as an environmental friendly corrosion inhibitor on the corrosion behavior of the sputter-deposited nanocrystalline Cr-10Zr-10W and amorphous Cr-9Ni-24W alloys in 0.5 M NaCl solution, open to air at 25±1ºC was studied using corrosion tests, corrosion mechanism and corrosion potential measurements. The following conclusions are drawn.

i. Sodium tungstate as a green corrosion inhibitor was found to be effective to decrease the corrosion rate (nearly one order of magnitude) of the chromium-based ternary alloys in 0.5 M NaCl solution.

ii. 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.

iii. Experimentally obtained values of the standard free energy of adsorption (ΔG^{o}_{ads}) of sodium tungstate for the Cr-10Zr-10W and Cr-9Ni-24W alloys were found to be -22.55 KJ/mol and -26.62 KJ/mol, respectively, suggested that the adsorption of the inhibitor onto alloy surfaces is due to physical adsorption, not by the chemical adsorption.

iv. Sodium tungstate acts as an anodic corrosion inhibitor for the two chromium-based ternary alloys in 0.5 M NaCl solution due to shifting of the corrosion potential of the alloys towards more positive direction with increasing the concentration of the inhibitor.

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