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Research Article

STUDY OF CORROSION INHIBITION MECHANISM OF SPUTTER-DEPOSITED W-42Cr-5Ni AND Cr-10Zr-10W ALLOYS BY SODIUM NITRITE AS GREEN INHIBITOR IN 0.5 M NaCl AND 1 M NaOH SOLUTIONS

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Abstract

The effect of different concentrations of sodium nitrite as a green corrosion inhibitor on the corrosion inhibition mechanism of the sputter-deposited ternary W-42Cr-5Ni and Cr-10Zr-10W alloys was studied in 0.5 M NaCl and 1 M NaOH solutions open to air at 25ºC using corrosion tests. The use of the sodium nitrite enhanced the corrosion resistance properties of both the alloys in 0.5 M NaCl and 1 M NaOH solutions. It is found that the sodium nitrite is strongly adsorbed on the surface of the sputter-deposited these two alloys by physical adsorption, not by the chemisorption. As a result the corrosion resistance property of the alloys was found to be significantly increased with increasing the concentration of the green corrosion inhibitor of sodium nitrite. The corrosion rates of the sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys were decreased with increasing the concentrations of sodium nitrite of 2400 ppm.

Key words: Green corrosion inhibitor; W-42Cr-5Ni alloy; Cr-10Zr-10W alloy; sodium nitrite; 0.5 M NaCl; 1 M NaOH

Introduction

Corrosion is an undesirable phenomenon which destroys the properties of metallic materials and shortens life of the materials (Bhattarai, 2010a). 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 corrosion phenomena is a subject of tremendous technological significances. The corrosion phenomena can be controlled using different techniques. A choice of particular corrosion control techniques for a given system is very difficult works for both the corrosion scientists as well as the corrosion engineers. 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 scientitsts and engineeres (Bhattarai, 2010a; Uhlig and Revie, 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 metallic materials exposed to that environment (Uhlig and Revie, 2008; Hackerman and Snaveley, 1984). A 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 such inhibitor to retard the corrosion rate of the materials is becoming one of the widely used corrosion control methods. The corrosion control practices by adding different environmental friendly green inorganic corrosion inhibitors such as nitrites, tungstates and molybdates. There are several types of such inhibitors designated are inorganic, organic, vapor−phase inhibitors and including organic as well as vapor-phase inhibitors.

Practical criteria for the selection of corrosion inhibitors from the variety of inorganic and organic compounds with inhibiting properties are not only their inhibition efficiency but also safety of use, economic constraints and compatibility with other chemicals in the system as well as the environmental concerns. In recent years, many alternative eco-friendly green corrosion inhibitors are developed, the range from inorganic (Kalyani and Rao, 2014; Gaun, 2007; Sribhurathy and Rejendran, 2012; Acharya *et al.*, 2013) to organic compounds (Acharya *et al.*, 2013; Sundaram *et al.*, 2013; Merest *et al.*, 2012; Afshari and Dehghanian, 2010; Zhang and Hua, 2009; Chandrasekar *et al.*, 2006; Bekkouch, 2003). A lot of natural products were used as the corrosion inhibitors for different metals and alloys in various environments (Sundaram *et al.*, 2013; Merest *et al.*, 2012; Afshari and Dehghanian, 2010; Chandrasekar *et al.*, 2006; Bekkouch, 2003; Negm *et al.*, 2013; Sriram *et al.*, 2014; Vasudha and Priya, 2014; Vastag *et al.*, 2001; Li and Lei, 2011; Mahmoud, 2007; Prabhu and Rao, 2013). Similary, inorganic compounds are also widely used as effective corrosion inhibitors to control the corrosion rate of metallic materials in corrosive environments. However, all of these inorganic corrosion inhibitors are not eco-friendly inhibitors. For example, chromate (VI) is reported as one of the most effective inhibitors, but it is toxic to human beings (Lei *et al.*, 2011; Ilevbare and Burstein, 2003; Twite and Bierwagen, 1998; Kendig and Buchheit, 2003). Therefore, nowadays Cr(VI) is not generally used as corrosion inhibitors. There is great interest in replacing chromates with effective and non-hazardous alternatives green corrosion inhibitors. Nitrites, molybdate, tungstate are now being increasingly used as eco-friendly green inorganic corrosion inhibitors, because of their low order of toxicity (Subedi *et al*., 2014; Eghbali *et al*., 2011; Zhao and Zuo, 2002; Celeste and Vieira, 2004; Saji and Thomas, 2007; Ali *et al.*, 2009; Refaey *et al.*, 2000; Pryor and Cohen, 1953; Lizlovs, 1976; Farr and Saremi, 1982; Mustafa and Dulal, 1997).

It was reported that the sputter-deposited amorphous or/and nanocrystalline ternary W-xCr-yNi (Bhattarai, 2009, 2010b; Bhattarai and Kharel, 2009-10; Kharel and Bhattarai, 2009) and W-xZr-yCr (Aryal and Bhattarai, 2010; Bhattarai, 2010c, 2011a, 2011b; Bhattarai and Aryal, 2011; Kumal and Bhattarai, 2010) alloys were spontaneously passivated showing significantly higher corrosion resistance than those of alloy-constituting elements in different aggressive solutions. In this context, the present work is focused to study the effects of the eco-friendly green corrosion inhibitor of sodium nitrite to control the corrosion of two sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys in 0.5 M NaCl and 1 M NaOH solutions open to air at 25ºC using corrosion tests and mechanism.

Materials and Methods

Sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys

X–ray diffractometer with CuKα radiation at $\theta - 2\theta$ mode and electron probe microanalysis were used to determine the composition and structure of the sputter deposit of the W-42Cr-5Ni and Cr-10Zr-10W alloys, , respectively (Bhattarai, 2009, 2010b and 2010c). Apparent grain size of the alloys was estimated using Scherrer's formula (Cullity, 1977).

Corrosion test and inhibition mechanism

In order to explain the corrosion inhibition mechanism for the corrosion control of the sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys using different concentrations of sodium nitrite as eco-friendly green corrosion inhibitor in 0.5 M NaCl and 1 M NaOH solutions open to air at 25ºC, the degree of surface coverage (Θ) and the standard free energy change of adsorption of the inhibitor onto the alloy surface were estimated using corrosion tests. The corrosion test of the W-42Cr-5Ni and Cr-10Zr-10W alloys was carried out in 0.5 M NaCl and 1 M NaOH solutions with different concentrations (i.e. 200, 400, 800, 1200, 1600 and 2400 ppm) of sodium nitrite. Before each corrosion tests, the alloy specimen was polished mechanically using silicon carbide paper having 1500 grit number in cyclohexane. The polished alloy specimens were rinsed with acetone and dried it in air to obtain reproducible results by removing oxide film of the alloy specimen surface. The weight loss method was used to estimate the average corrosion rate of the alloys using the following equation 1(Bhattarai, 2010a). The estimation of the average corrosion rate was carried out two times or more so as to obtained delimited results.

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

Where, Δw is the weight loss of the 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.

The inhibition efficiency (IE_{CR}) and the degree of surface coverage (θ) of the inhibitor molecule adsorbed on the alloy surface (Hegazy *et al.*, 2012; Negm *et al.*, 2012; Narváez *et al.*, 2005) were estimated using following equations 2 and 3, respectively, where, $CR_{(unhib.)}$ and $CR_{(inhib.)}$ are the corrosion rates in absence and presence of the corrosion inhibitors, respectively.

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

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

Corrosion inhibition mechanism was studied using Langmuir adsorption model. The Langmuir adsorption isothermal 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.

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

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^{o}_{ads}) using equations 5 and 6 (Cases and Villieras, 1992).

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

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

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

Results and Discussion

Characterization of the alloys

Two sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys were used to carry out this work. The instrument used and conditions subjected for the sputter deposition of these two alloys were same as those described elsewhere (Bhattarai, 2009, 2010b and 2010c). The composition, apparent grain size and structure of the alloys were analyzed by EPMA and XRD, respectively. The results of the characterization of these alloys including alloy-constituting elements are summarized in Table 1. Alloy compositions hereafter are expressed in atomic percentage (at %). It was found that the apparent grain size of the W-42Cr-5Ni, Cr-10Zr-10W alloys including tungsten, chromium, nickel and zirconium metals was found to be about 3.5, 29, 20, 40, 19 and 24 nm, respectively. Accordingly, the W-42Cr-5Ni and Cr-10Zr-10W alloys were characterized as the mixture of nanocrystalline and amorphous, and a nanocrystalline structures having the apparent grain size of 3.5 nm (Bhattarai 2009 and 2010b) and 29 nm (Bhattarai 2010c and Bhattarai 2011a), respectively.

Inhibition effect on the corrosion

The effect of the green corrosion inhibitor of sodium nitrite on the corrosion rate of the sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys was estimated after immersion for 240 h in 0.5 M NaCl and 1 M NaOH solutions open to air at 25ºC in absence and presence of different concentrations (i.e., 200-2400 ppm) of sodium nitrite. The corrosion rate of the W-42Cr-5Ni and Cr-10Zr-10W alloys is decreased with increasing the concentrations of sodium nitrite up to 1600 ppm and then the corrosion rates become steady between 1600-2400 ppm of nitrite as depicted in Figs 1(a) and 1(b), respectively. The corrosion rates of both alloys were found to be lowest in 2400 ppm sodium nitrite in both solutions as clearly shown in Figs 1(a) and 1(b). It is assumed that sodium nitrite concentration ranges from 1200 to 2400 ppm is sufficient to decrease the corrosion rate of the alloys in 0.5 M NaCl and 1 M NaOH solutions. However, the trend of decrease of the corrosion rate with increasing the concentrations of the sodium nitrite is not same. As a result the corrosion rate of the sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys in 1 M NaOH solution was found higher than in 0.5 M NaCl solution at 25ºC. This is probably due to show high corrosion rates of both the alloys in 1 M NaOH than in 0.5 M NaCl solution only in absence of sodium nitrite as shown in Figs 1(a) and 1(b). These results revealed that the use of sodium nitrite as a green corrosion inhibitor enhanced to increase the corrosion resistance properties of the W-xCr-yNi and Cr-xZr-yW alloys in 0.5 M NaCl and 1 M NaOH solutions.

Corrosion inhibition efficiency

The inhibition efficiency of sodium nitrite in 0.5 M NaCl and 1 M NaOH solutions for the W-42Cr-5Ni and Cr-10Zr-10W alloys is increased with increasing the inhibitor concentrations as shown in Figs 2 (a) and 2 (b), respectively. Similarly the maximum inhibition efficiency of about 80-85 % was obtained between 1200 to 2400 ppm of sodium nitrite in 0.5 M NaCl solution whereas it was found to be between 65 to 70 % in 1 M NaOH solution for the W-42Cr-5Ni alloy as shown in Fig. 2 (a). Similalry, the maximum inhibition efficiency of about 80-92 % was obtained between 1200 to 2400 ppm of sodium nitrite in 0.5 M NaCl solution whereas it was found to be around 60-75 % in 1 M NaOH solution for the Cr-10Zr-10W alloy as shown in Fig. 2 (b). It is found that the most efficient corrosion inhibitor concentration of sodium nitrite is found to be at 2400 ppm for both the alloys in 0.5 M NaCl solution at 25ºC

Table 1: Chemical composition, apparent grain size and structure of the sputter-deposited W-42Cr-5Ni and Cr-10Zr-10W alloys including alloy-constituting tungsten, chromium, zirconium and nickel.

Metals & Alloys	Tungsten Content (at %)	Chromium Content (at %)	Zirconium Content (at %)	Nickel Content (at %)	Apparent Grain Size (nm	Structure*
Tungsten	100	-	-	-	20	Nanocryst.
Chromium	-	100	-	-	40	Nanocryst.
W-42Cr-5Ni	53.53	41.54	-	4.93	3.5	Nanocryst. + Amor.
Cr-10Zr-10W	10.08	79.98	9.94	-	29	Nanocryst.
Zirconium	-	-	100	-	24	Nanocryst.
Nickel	-	-	-	100	19	Nanocryst.

** Nanocryst. = Nanocrystalline and Amor. = Amorphous*

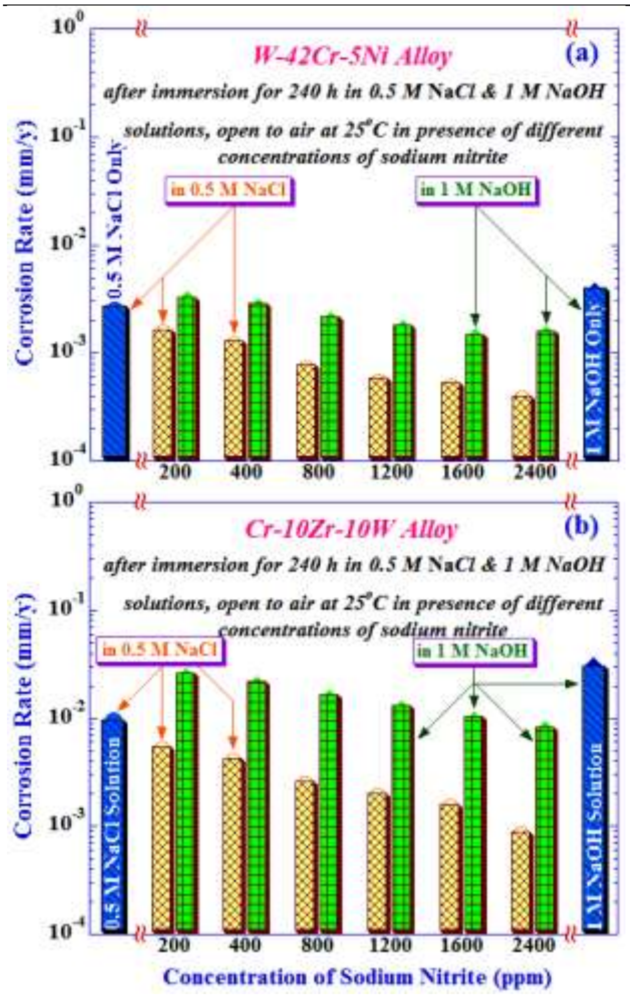

Fig. 1: Changes in the corrosion rates of the sputter-deposited (a) W-42Cr-5Ni and (b) Cr-10Zr-10W alloys after immersion for 240 hours in 0.5 M NaCl solution open to air at 25ºC, as a function of the concentration sodium nitrite as a green corrosion inhibitor.

The inhibition action of the tested sodium nitrite as green corrosion inhibitor was found to obey the Langmuir isotherm model of adsorption. Consequently, the increase in the percentage corrosion inhibition efficiency with increasing inhibitor concentrations indicated that the sodium nitrite acting as adsorption inhibitor to decrease the corrosion rate of the W-xCr-yNi and Cr-xZr-yW alloys in near neutral 0.5 M NaCl and alkaline 1 M NaOH solutions. The results revealed that the percentage corrosion inhibition efficiency was increased with increasing the inhibitor concentration in 0.5 M NaCl and 1 M NaOH solutions.

Corrosion inhibition mechanism

The Langmuir adsorption isotherm model was performed to have more insights into the mechanism of corrosion inhibition of sodium nitrite on the W-42Cr-5Ni and Cr-10Zr-10W alloys in 0.5 M NaCl and 1 M NaOH solutions open to air at 25ºC. In order to obtain adsorption isotherm to explain the corrosion inhibition mechanism of sodium nitrite for the W-xCr-yNi and Cr-xZr-yW alloys, the ϴ value was estimated from the weight loss measurement at different concentrations of sodium nitrite in 0.5 M NaCl and 1 M NaOH solutions at 25ºC. The process of inhibitor adsorption on the surface of the alloys can be described by different isotherms, from which Langmuir model is simplest and based on the assumption that all adsorption sites are equivalent and the particle binding occurs independently from nearby sites being occupied or not (Satapathy et al., 2009) as expressed in equation (4) above.

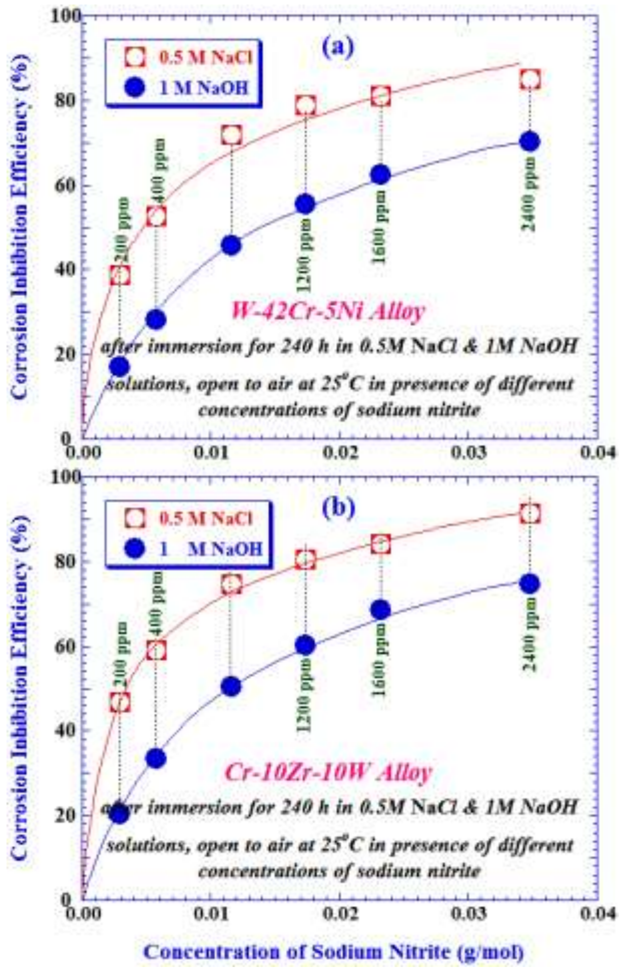

Fig. 2: Changes in the corrosion inhibition efficiency on the sputter-deposited ternary (a) W-42Cr-5Ni and (b) Cr-10Zr-10W alloys after immersion for 240 hours in 0.5 M NaCl solution open to air at 25ºC, as a function of sodium nitrite concentration.

Figures 3 (a) and 3 (b) show the relationship between C/ϴ and C for the corrosion inhibitor of sodium nitrite in 0.5 M NaCl and 1 M NaOH solutions for W-42Cr-5Ni and Cr-10Zr-10W alloys, respectively. The linear correlation coefficient (R^2) and the slope of the straight line for the alloys in 0.5 M NaCl and 1 M NaOH solutions 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 nitrite in 0.5 M NaCl and 1 M NaOH solutions.

It is meaningful for mentioning here the fact that there is no interaction between the adsorbed corrosion inhibitor molecules, the energy of adsorption is independent on the ϴ, the alloy surfaces contain a fixed number of adsorption sites and each site holds one adsorbed species according to the Langmuir isotherm model (Cases and Villieras, 1992). The standard free energy of adsorption (ΔG°ads), which can characterize the interaction of adsorption molecules of the inhibitors and alloy surfaces, was calculated using equations (4) and (5) as discussed above.

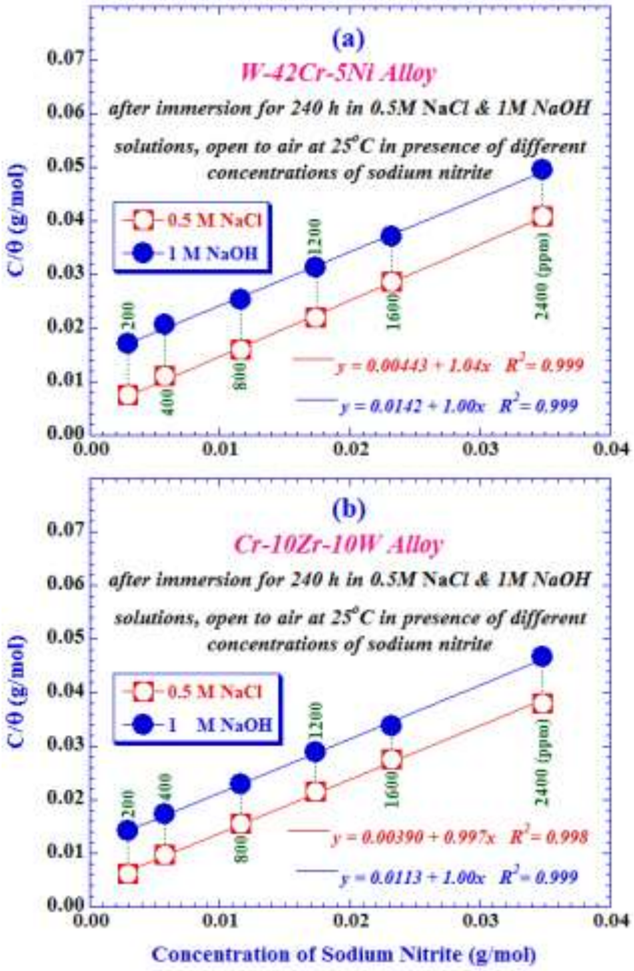

Fig. 3: Langmuir's adsorption plots for the sputter-deposited (a) W-42Cr-5Ni and (b) Cr-10Zr-10W alloys in 0.5 M NaCl and 1 M NaOH solutions in presence of different concentrations of sodium nitrite as a corrosion inhibitor.

In literature, it was reported that the high K (= 1/K_{ads}) value (greater than 100 M^{-1}) attributes to stronger and more stable adsorbed layer formation on the metals or/and alloys surfaces (Dahmani *et al*., 2010; Ahmada *et al.*, 2010). It was reported that the ΔG^{o}_{ads} values of metals or alloys around -20 KJ/mole or lower indicate adsorption of inhibitors onto metals or alloys surface with electrostatic interaction is due to physical adsorption, while those around or higher (more negative) than -40 KJ/mole involve charge sharing between inhibitor molecules and metals or alloys surfaces is of chemisorption (Khaleda, 2010; Zhang and Hua, 2010). 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 metals or alloys surfaces.

It was found that the ΔG^{o}_{ads} values for sodium nitrite as corrosion inhibitor in 0.5 M NaCl and 1 M NaOH solutions at 25ºC for the W-42Cr-5Ni alloy were estimated -23.38 and -20.49 KJ/mole, respectively and for the Cr-10Zr-10W alloy estimated -23.70 KJ/mole and -22.55 KJ/mole, respectively, which are consistent with literatures. Hence authenticates physical adsorption of the inhibitors on the surface of both the W-42Cr-5Ni and the Cr-10Zr-10W alloys. This implies that the corrosion inhibitors of sodium nitrite adhere on the surface of the sputter-deposited alloys and so give more efficient inhibition effects to decrease the corrosion rate of the alloys in both the 0.5 M NaCl as well as 1 M NaOH solutions open to air at 25ºC.

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