

## Roles of Alloying Elements on the Passivity of W-xCr-yNi Alloys in Aggressive Environments

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### Abstract

The role of alloying elements on the passivity of the sputter-deposited amorphous or nanocrystalline W-xCr-yNi alloys in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C was studied by corrosion tests and electrochemical measurements. All the examined sputter-deposited W-(42-75)Cr-(4-15)Ni alloys, which are composed of either amorphous or nanocrystalline single phase solid solution, showed higher corrosion resistance than those of alloying elements (i.e. tungsten, chromium and nickel) in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions. All the alloys used in this study showed higher corrosion resistance than those of the alloying elements in the aggressive environments. Anodic current densities of the alloys decreased with increasing chromium contents and became lower than that of chromium. Transpassive dissolution of chromium was significantly enhanced by the simultaneous additions of tungsten, chromium and nickel in aggressive environments of 12 M HCl, 0.5 M NaCl and 1 M NaOH. Hence the synergistic effect of chromium addition in enhancing the corrosion resistance of the W-xCr-yNi alloys was clearly observed from the present study.

**Key words:** W-xCr-yNi alloys, sputter deposition, corrosion resistance, 12 M HCl, 0.5 M NaCl, 1 M NaOH

### Introduction

The chemically homogeneous single-phase nature of amorphous or/and nanocrystalline alloys is generally responsible for their high corrosion resistance owing to the formation of uniform protective passive films which are able to separate bulk of alloys from aggressive environments. In recent years, the use of the sputter deposition technique is becoming a quite suitable for tailoring of varieties of amorphous or/and nanocrystalline corrosion-resistant alloys, because the sputter deposits are considered to be homogeneous alloys from corrosion point of view.

Tungsten, chromium and nickel are known as effective alloying elements in enhancing the corrosion resistance properties of alloys in aggressive environments. It has been reported that the sputter-deposited amorphous or/and nanocrystalline tungsten-based W-xTi (Bhattarai 2009a, 2009b, Bhattarai *et al.* 1995, Sharmah *et al.* 2007), W-xNi (Bhattarai 2001, Sah & Bhattarai 2008/2009), W-xNb

(Bhattarai 2011a, Bhattarai *et al.* 1998a, 1998b, Jha & Bhattarai 2008), W-xCr (Bhattarai *et al.* 1998c, Bhattarai & Hashimoto 1998, Basnet and Bhattarai 2010), W-xTa (Bhattarai 2011b, 2012a, Bhattarai & Baral 2013, Bhattarai *et al.* 1998d), W-xMo (Bhattarai 2006, 2012b, Khadka & Bhattarai 2010), W-xZr (Bhattarai *et al.* 1997, Shrestha & Bhattarai 2010), W-xZr-yCr (Aryal & Bhattarai 2010, 2011, Bhattarai 2010a, 2011c, 2011d, Kumal & Bhattarai 2011) and W-xCr-yNi (Bhattarai 2009c, 2010b, Bhattarai & Kharel 2009-10, Kharel & Bhattarai 2009) alloys were spontaneously passivated showing significantly higher corrosion resistance than those of alloy-constituting elements in aggressive solutions. The significant improvement of the corrosion resistance properties of these alloys is attributed to the formation of double oxyhydroxide passive films containing both tungsten and alloy-constituting metal cations.

On the other hand, chromium is one of the corrosion-resistant elements in aggressive environments,

although it suffers transpassive dissolution by anodic polarization in chloride containing solutions. The alloying of chromium with zirconium (Kim *et al.* 1993a, Li *et al.* 1997a), titanium (Kim *et al.* 1993b, Li *et al.* 1997b), niobium and tantalum (Kim *et al.* 1994), molybdenum (Park *et al.* 1995) and tungsten (Bhattacharai *et al.* 1998c, Bhattacharai & Hashimoto 1998) greatly improved the corrosion resistance of the alloys in acidic solutions. In particular, it has been reported that only 10 at% of tungsten addition was enough to cause spontaneous passivation of the nanocrystalline W-xCr alloys in 12 M HCl solution and these alloys showed lower corrosion rates than those of tungsten and chromium (Bhattacharai *et al.* 1998c). Furthermore, chromium is proved as one of the most effective alloying elements to provide a high passivating ability particularly for showing high pitting corrosion resistance of the sputter-deposited binary Cr-Zr alloys (Kim *et al.* 1993a, Li *et al.* 1997a) and Mo-Cr alloys (Park *et al.* 1995) in hydrochloric acid solutions. Similarly, a series of the sputter-deposited chromium-metalloid alloys exhibited superior corrosion resistance properties compared to chromium metal in HCl solutions (Moffat *et al.* 1995).

Furthermore, it has been reported that the sputter-deposited amorphous or/and nanocrystalline W-Ni alloys were passivated spontaneously and tungsten addition greatly enhances the corrosion resistance properties of nickel in 12 M HCl (Bhattacharai 2001). It is noteworthy for mentioning here that both chromium (Pourbaix 1974a) and nickel (Pourbaix 1974b) metals are very corrodible in acidic or very alkaline (>13 pH) solutions containing oxidizing agents. However, both chromium and nickel metals do not generally corrode in alkaline solutions like 1 M NaOH, mostly due to the formation of chromium hydroxide/oxides (Pourbaix 1974a) and nickel hydroxide/oxides (Pourbaix 1974b), respectively. On the other hand, tungsten metal is actively dissolved even in slightly alkaline solution as well as in solution having pH of 4 or high (Pourbaix 1974c). Accordingly, it might be expected that effects of tungsten, chromium and nickel additions enhance in the corrosion resistance properties of the sputter-deposited amorphous or nanocrystalline W-xCr-(4-15)Ni alloys in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions.

In this context, the present research is aimed to clarify the roles of tungsten, zirconium and chromium

additions on the passivity of the ternary W-xCr-(4-15)Ni alloys in different aggressive environments, that is, in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions.

## Methodology

The sputter-deposited ternary W-xCr-yNi alloys containing 42-75 chromium and 4-15 nickel were characterized as single-phase solid solutions of amorphous or/and nanocrystalline structures having apparent grain size ranges from 1.5 to 10.2 nm (Bhattacharai 2009c, 2010b, Bhattacharai & Kharel 2009-10, Kharel & Bhattacharai 2009). The composition of the sputter-deposited W-xCr-yNi alloys hereafter is denoted in atomic percentage (at%).

Prior to corrosion tests and electrochemical measurements, the surface of the sputter-deposited ternary W-xCr-yNi alloy specimens was polished mechanically with silicon carbide paper grit No. 1500 in cyclohexane, rinsed with acetone and dried in air. Corrosion rates of the W-xCr-yNi alloys including tungsten, chromium and nickel metals were estimated from the weight loss after immersion for 168 h in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C. Potentiodynamic anodic polarization curves were measured after immersion for 30 minutes in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions at 30°C when the open circuit potential became almost steady. The scan rate of the polarized potential was adjusted at 1 mV.s<sup>-1</sup>. A platinum mesh and a saturated calomel electrode (SCE) were used as counter and reference electrodes, respectively. All the potential given in this paper are relative to SCE.

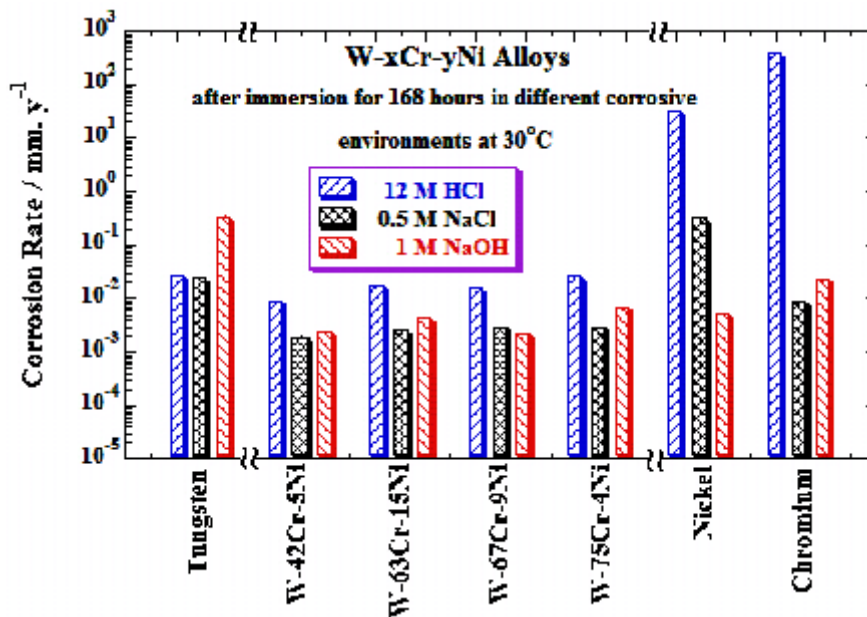
## Results and Discussion

Figure 1 shows corrosion rates of the sputter-deposited amorphous or nanocrystalline W-(42-75)Cr-(4-15)Ni alloys after immersion for 168 h in different aggressive environments at 30°C. The corrosion rates of the sputter-deposited tungsten, chromium and nickel metals are also shown for comparison. The additions of 42-75 at% chromium and 4-15 at% nickel in the W-xCr-yNi alloys significantly enhanced the corrosion resistance properties of the alloys in aggressive 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions. As a result, the corrosion rates of the W-xCr-yNi alloys, which are composed of either amorphous or nanocrystalline phase, are lower than those of alloying elements in the corrosive environments. Furthermore, all the examined alloys

showed higher corrosion rates in 12 M HCl solution than those in 0.5 M NaCl and 1 M NaOH solutions. It is meaningful for mentioning here that the corrosion rates of these ternary alloys were found to be lower than those of the sputter-deposited binary W-(42-75)Cr (Basnet and Bhattacharai 2010, Bhattacharai *et al.* 1998c) and W-(4-15)Ni (Bhattacharai 2001, Sah & Bhattacharai 2008/2009) alloys in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions.

In particular, corrosion rates of the W-xCr-(4-15)Ni alloys containing 42-75 at% chromium were found to be lower than that of the sputter-deposited W-15Ni alloys in 12 M HCl solution (Bhattacharai 2009c). Consequently, a small addition of nickel was found to be detrimental for the corrosion resistance properties of the W-Cr-Ni alloys in 12 M HCl solution (Bhattacharai 2009c). On the other hand, the corrosion rates of tungsten, chromium and nickel are about  $2.5 \times 10^{-2}$  mm.y<sup>-1</sup>,  $7.4 \times 10^{-3}$  mm.y<sup>-1</sup> and  $3.0 \times 10^{-1}$  mm.y<sup>-1</sup>,

respectively, in 0.5 M NaCl solution. It is noteworthy to mention here that corrosion rate of the sputter-deposited binary W-(4-15)Ni alloys was found to be in the range of  $1.95 \times 10^{-2}$ - $2.12 \times 10^{-2}$  mm.y<sup>-1</sup> in 0.5 M NaCl solution (Sah & Bhattacharai 2008/2009). The corrosion rates of the W-Cr-(4-15)Ni alloys containing 42-75 at% chromium were about one order of magnitude lower than those of the sputter-deposited W-(7-15)Ni alloys and tungsten, and even lower than that of the sputter-deposited chromium (Bhattacharai & Kharel 2009-10). These results clearly revealed that the addition of chromium to the W-(4-15)Ni alloys is effective in enhancing the corrosion resistance of the sputter-deposited W-Cr-(4-15)Ni alloys in 0.5 M NaCl solution. Consequently, the addition of 42-75 at% chromium content to the sputter-deposited binary W-(4-15)Ni alloys is very effective in enhancing the corrosion resistance properties of the sputter-deposited W-Cr-(4-15)Ni alloys in neutral 0.5 M NaCl solution.



**Fig. 1.** Changes of corrosion rates of the sputter-deposited W-xCr-yNi alloys including tungsten, chromium and nickel in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C

Similarly, the corrosion rates of the sputter-deposited binary W-(4-15)Ni alloys were found to be in the range of about  $2.1 \times 10^{-2}$ - $3.0 \times 10^{-1}$  mm.y<sup>-1</sup> in 1 M NaOH solution (Kharel & Bhattacharai 2009). However, the corrosion rates of the sputter-deposited W-Cr-(4-15)Ni alloys containing 42-75 at% chromium are about two orders of magnitude lower than those of the sputter-

deposited W-(7-9)Ni alloys and tungsten, and about one order of magnitude lower than those of sputter-deposited W-(4-15)Ni alloys as well as chromium metal in 1 M NaOH solution from the present study. Consequently, the addition of 42-75 at% chromium to the sputter-deposited binary W-(4-15)Ni alloys is very effective in enhancing the corrosion resistance of

the W-Cr-(4-15)Ni alloys in 1 M NaOH solution. From above discussion, it can be concluded that the simultaneous additions of tungsten and chromium with small amounts of nickel containing 4-15 at% enhance synergistically the corrosion resistance properties of the sputter-deposited ternary W-xCr-yNi alloys containing 42-75 at% chromium in aggressive 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions.

Electrochemical measurements were carried out for a better understanding of the high corrosion resistance properties of the sputter-deposited amorphous or/and nanocrystalline W-xCr-yNi alloys in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C. Figures 2-4 show the changes in the corrosion rates and the open circuit corrosion potentials of the alloys in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions in the given conditions, as a function of alloy chromium content. The open circuit corrosion potential of the alloys in 12 M HCl solution is shifted to more negative direction with increasing the chromium content in the ternary W-xCr-yNi alloys (Fig. 2), while the open circuit corrosion potential of all the examined alloys in 0.5 M NaCl (Fig. 3) and 1 M NaOH (Fig. 4) solutions is shifted to more positive direction with increasing the chromium content in the alloys. In particular, the sputter-deposited W-xCr-yNi alloys containing 63-75 at% chromium showed the most noble open circuit corrosion potential than those of W-42Cr-5Ni alloy, tungsten and chromium metals both in 0.5 M NaCl (Fig. 3) and in 1 M NaOH (Fig. 4) solutions. These results revealed that high corrosion resistance of the ternary W-xCr-yNi alloys is mostly due to the ennoblement of the open circuit corrosion potential of the alloys in 0.5 M NaCl and 1 M NaOH solutions at 30°C. Consequently, the sputter-deposited W-xCr-yNi alloys containing adequate amounts of tungsten and chromium showed the more stable passivity and showed higher corrosion resistance than those of alloy-constituting elements (i.e., tungsten, chromium and nickel) in 0.5 M NaCl and 1 M NaOH solutions.

On the other hand, the open circuit corrosion potentials of all the examined W-xCr-yNi alloys in 12 M HCl are located between the open circuit corrosion potentials of tungsten and chromium, but mostly very close to that of tungsten as shown in Fig. 2. It is meaningful to mention here that the open circuit corrosion potential of the W-xCr-yNi alloys containing 42-67 at% chromium are located very close to the

passive regions of the chromium metal and active regions of nickel metal as shown in Fig. 5. Accordingly, the additions of chromium and tungsten enhance the corrosion resistance properties of the sputter-deposited ternary W-xCr-yNi alloys and such a beneficial effect of nickel metal cannot be observed as shown in Fig. 1.

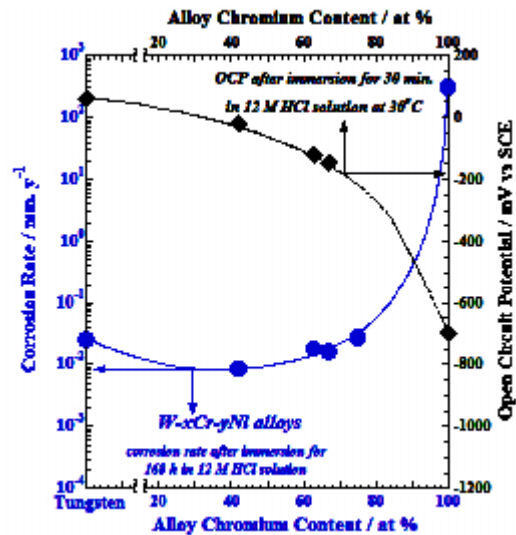


Fig. 2. Changes in corrosion rates and open circuit corrosion potentials of the sputter-deposited W-xCr-yNi alloys including tungsten and chromium in 12 M HCl solution open to air at 30°C

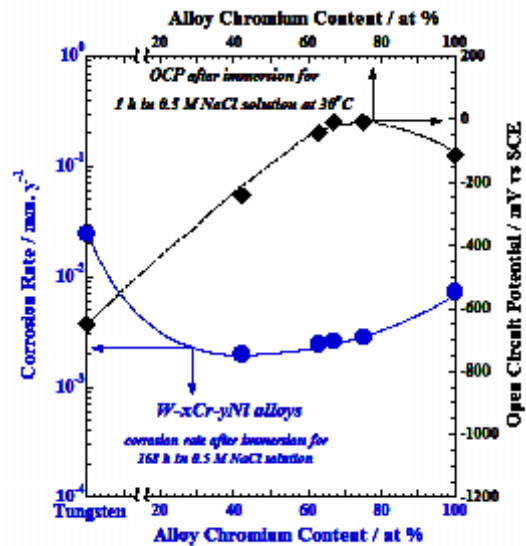


Fig. 3. Changes in corrosion rates and open circuit corrosion potentials of the sputter-deposited W-xCr-yNi alloys including tungsten and chromium in 0.5 M NaCl solution open to air at 30°C



Figures 5, 6 and 7 show the potentiodynamic anodic polarization curves for the ternary W-xCr-yNi alloys after immersion for 30 minutes in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions, respectively, at 30°C to determine the roles of tungsten, chromium and nickel additions on the anodic passivity of the alloys in the aggressive environments. The anodic polarization curves for the sputter-deposited tungsten, chromium and nickel metals are also shown for comparison.

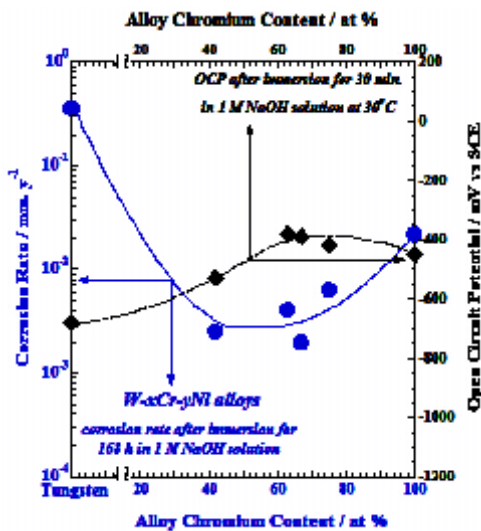


Fig. 4. Changes in corrosion rates and open circuit corrosion potentials of the sputter-deposited W-xCr-yNi alloys including tungsten and chromium in 1 M NaOH solution open to air at 30°C

All the examined W-xCr-yNi alloys as well as tungsten metal are spontaneously passivated in 12 M HCl in contrast to chromium and nickel metals those show the active-passive transitions as shown in Fig. 5. Chromium metal shows the transpassive dissolution at about 0.80 V vs SCE mostly due to the formation of hexavalent chromium ion in 12 M HCl solution. The anodic current densities of the sputter-deposited W-xCr-yNi alloys decrease with increasing the chromium content in the alloys. The W-75Cr-4Ni alloy shows the lowest anodic current density among the examined alloys in this study. It is noteworthy to mention that the anodic current densities of the alloys are significantly lower than those of the alloying elements (i.e., tungsten, chromium and nickel). Accordingly, the protective quality of the anodic passive films formed on the sputter-deposited W-xCr-yNi alloys is better than those of alloying elements even in very aggressive environment of 12 M HCl solution at 30°C.

Similarly, all the examined W-xCr-yNi alloys as well as tungsten, chromium and nickel metals are spontaneously passivated in both 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C as shown in Figs 6 and 7. However, all the alloys including nickel and chromium metals showed the transpassive dissolution at higher anodic potential regions. In general, the transpassive dissolution potential of all the examined alloys in 0.5 M NaCl solution was found to be shifted to more positive or noble direction than in 1 M NaOH solution. The anodic current density of the alloys is decreased with increasing the chromium content in the alloys in both 0.5 M NaCl and 1 M NaOH solutions.

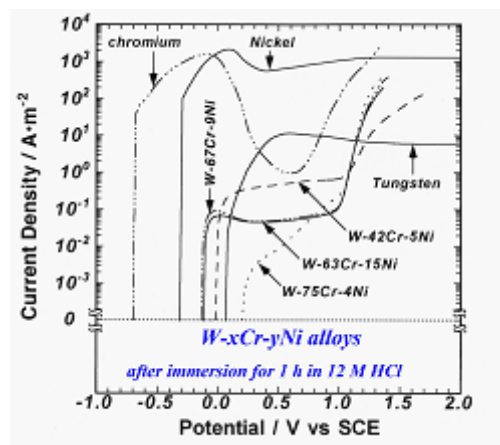


Fig. 5. Potentiodynamic polarization curves for the sputter-deposited ternary W-xCr-yNi alloys including sputter-deposited tungsten, chromium and nickel metals in 12 M HCl solution open to air at 30°C

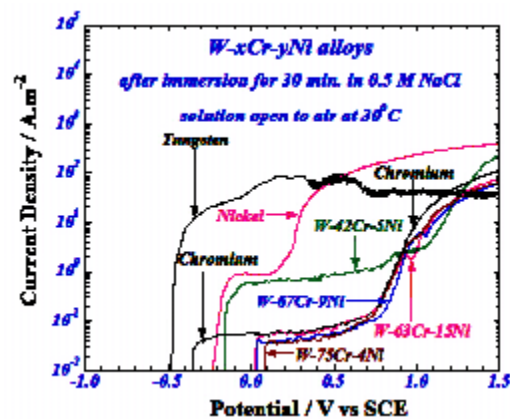


Fig. 6. Potentiodynamic polarization curves for the sputter-deposited ternary W-xCr-yNi alloys including sputter-deposited tungsten, chromium and nickel metals in 0.5 M NaCl solution open to air at 30°C

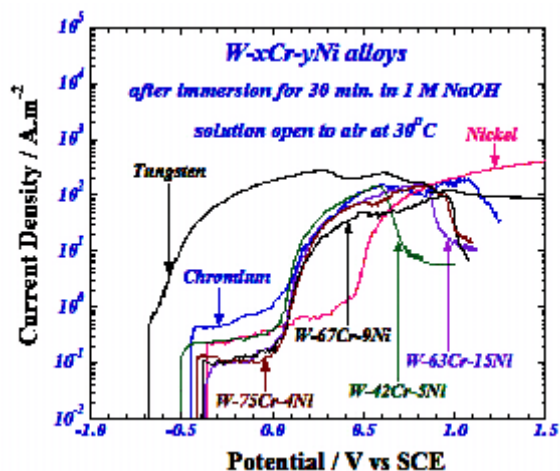


Fig. 7. Potentiodynamic polarization curves for the sputter-deposited ternary W-xCr-yNi alloys including sputter-deposited tungsten, chromium and nickel metals in 1 M NaOH solution open to air at 30°C

In particular, the current density of the W-42Cr-5Ni alloy is slightly higher than those of other W-xCr-yNi alloys in both 0.5 M NaCl and 1 M NaOH solutions. These results revealed that the protective quality of the anodic passive films formed on the sputter-deposited W-xCr-yNi alloys is better than those films formed on the alloying elements in both 0.5 M NaCl and 1 M NaOH solutions. The open circuit corrosion potential of all the examined ternary W-xCr-yNi alloys in both 0.5 M NaCl and 1 M NaOH solutions is shifted to more positive or noble direction than those of the sputter-deposited tungsten and chromium. These results are in support of the results of corrosion studies as shown in Fig. 1.

In summary, the synergistic effects of the simultaneous additions of tungsten, chromium and nickel in enhancing the corrosion resistance of the amorphous or/and nanocrystalline W-xCr-yNi alloys containing 42-75 at% chromium and 4-15 at% nickel is clearly observed in 12 M HCl, 0.5 M NaCl and 1 M NaOH solutions open to air at 30°C and hence all the examined sputter-deposited ternary W-xCr-yNi alloys showed higher corrosion resistance than those of alloy-constituting elements (i.e., tungsten, chromium and nickel). The simultaneous additions of chromium and nickel to tungsten metal provided outstanding improvement in the protective quality of the anodic passive film in comparison with the single addition of tungsten or chromium on zirconium and hence both tungsten and chromium act synergistically in

enhancing the transpassive dissolution of chromium metal in aggressive environments. The open circuit corrosion potentials of the W-63Cr-15Ni, W-67Cr-9Ni and W-75Cr-4Ni alloys are shifted to more noble (or positive) direction than those of all alloy-constituting elements after immersion for 30 minutes in all corrosive environments used in the present study.

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