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Corrosion resistance of CrN thin films produced by dc magnetron sputtering

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A R T I C L E I N F O

ABSTRACT

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Keywords: Corrosion EIS Pressure Tafel curves XRD In this study, the electrochemical behavior of chromium nitride (CrN) coatings deposited on two steel substrates, AISI 304 and AISI 1440, was investigated. The CrN coatings were prepared using a reactive d.c. magnetron sputtering deposition technique at two different pressures (P1 = 0.4 Pa and P2 = 4 Pa) with a mixture of N₂-Ar (1.5-10). The microstructure and crystallinity of the CrN coatings were investigated using X-ray diffraction. The aqueous corrosion behavior of the coatings was evaluated using two methods. The polarization resistance (Tafel curves) and electrochemical impedance spectra (EIS) in a saline (3.5% NaCl solution) environment were measured in terms of the open-circuit potentials and polarization resistance (R_p). The results indicated that the CrN coatings present better corrosion resistance and R_p values than do the uncoated steel substrates, especially for the coatings produced on the AISI 304 substrates, which exhibited a strong enhancement in the corrosion resistance. Furthermore, better behavior was observed for the coatings produced at lower pressures (0.4 Pa) than those grown at 4 Pa.

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1. Introduction

Ceramic coatings composed of transition metal nitrides, such as TiN, TiAlN, CrN and NbN, have been widely applied in industrial applications because they provide wear protection, heat and corrosion resistance, good adhesion and a high level of hardness [1,2]. In addition to TiN, CrN is another popular coating because it is a tough and superior coating for wear and corrosion resistance applications, especially for soft substrates used in plastic extrusion [3] and die casting molds [4,5]. CrN coatings as monolayers and multilayers combined with other compounds have been produced using different techniques, and the most commonly used techniques for producing these coatings are cathodic vacuum arc and magnetron sputtering [6–8]. Moreover, one of the most interesting applications for CrN is as a corrosion-resistant protective coating.

The corrosion of hard coatings on steel produced using plasmaassisted physical vapor deposition usually takes a localized form due to the establishment of an electropotential difference between the coating material and the less noble steel substrate [9]. Based upon this electrochemical corrosion characteristic, the factors that affect the corrosion resistance of coatings, such as CrN, on steel have been investigated for many years with respect to pit initiation and further development at through-coating defects in PVD (physical vapor deposition) hard coating systems [10,11]. The primary advantage for depositing CrN films is the low deposition temperature, and this characteristic makes it suitable for deposition onto temperature-sensitive materials and metals with low melting points [12,13].

However, it is well-known that the deposition parameters, such as the pressure and substrate material, have a considerable influence on the film properties; subsequently, the different functions of a component are performed by different layers, including the substrate, the substrate-coating interface and the coating layer. The purpose of a coating is generally related to wear resistance, protection against corrosion, thermal and electrical insulation, and the optical appearance of the work-piece. The substrate-coating interface is important for adhesion and providing a diffusion barrier, whereas the substrate can impart strength, stiffness and weight [14]. Although there are many reports on studies of corrosion resistance in CrN coatings [15-17], we are interested in investigating the differences in the behavior of coatings grown on two widely used steels that contain different percentages of Cr under two values of a N₂ flux. There are many studies in the literature concerning the influence of the substrate on the coating characteristics; nevertheless, we will focus our attention on the differences in the structural and corrosion resistance behavior of coatings when they are grown on two different Cr-based steels, which provides a possible synergistic relationship between the coating-substrate. Furthermore, it was demonstrated that although the substrate is highly suitable for growing the coatings, other deposition conditions, such as the working pressure, can influence the corrosion resistance of

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the coatings. Therefore, to enhance the behavior of the coatings, it is necessary to combine the investigation of the effects of several parameters, such as the substrate, pressure as in this study, and other parameters, such as the temperature, bias voltage and concentration of gases.

In this work, an investigation of the corrosion resistance of CrN coatings produced by d.c. magnetron sputtering on two steel substrates at two different working pressure is presented. The corrosion resistance was analyzed using polarization resistance (Tafel curves) and electrochemical impedance spectroscopy (EIS) measurements.

2. Experimental setup

Two substrates were selected to produce the coatings. The austenitic AISI 304 steel has one of the highest corrosion resistances compared to other Cr-Ni (carbon stabilized) based steels when it is exposed to several harmful environments [18]. A common misconception concerning stainless steel is that it is not affected by corrosion. While misleading, the phenomenal success of the metal makes this common belief understandable. When held in the temperature range between 800 and 1650 °F, austenitic stainless steels may undergo a change that renders them susceptible to intergranular corrosion when exposed to a number of corrosives, including certain corrosives that may otherwise only have a slight effect on them. One of New York City's most impressive landmarks is the stainless steel-clad peak of the Chrysler Building, which was built in 1930 of 302 stainless steel. A recent inspection of this landmark revealed no signs of corrosion or loss of thickness. The tallest manmade monument in the US, the St. Louis Arch, is entirely clad in 304 stainless steel plates; nevertheless, all austenitic stainless steels contain a small amount of carbon. At extremely high temperatures, such as those experienced during welding, the carbon forces the local chrome to form chromium carbide around it, which consequently starves the adjacent areas of the chrome required for its own corrosion protection. When welding, it is recommended to consider using low-carbon stainless steel, such as 304 and 316 [19,20].

The AISI-SAE 4140 steel is a Cr-Mo alloyed steel that has high stability up to 400 °C and is suitable for resisting stress and torsion [21]. The dimensions of the sample were 1.25 cm in diameter and 4 mm in thickness. The samples were polished using abrasive silicon carbide paper, and they subsequently were thoroughly cleaned using an ultrasonic cube in acetone for 15 min to eliminate oil, dust and any contamination. A d.c. magnetron sputtering system placed in a class 1000 clean-room was employed, and the base vacuum pressure was 5×10^{-5} Pa. The system is composed of multisource equipment (ATC 1500) from AJA International. The coatings were grown using the following conditions: a target of Cr (99.99%), room temperature (RT), -300 V as a bias voltage, inter-electrode distance of 10 cm, laboratory temperature of 24 °C and a relative humidity of 55%. Furthermore, two working pressures (P1 = 0.4 Pa and P2 = 4 Pa), a gas mixture of N₂-Ar (1.5-10 sccm) and a power of 8 W/m^2 for 90 min were employed.

The XRD analysis was performed using a Rigaku Ultima III with Cu K α radiation, which had a wavelength of λ = 0.1540 nm and used 40 kV with 44 mA for the source power. The Rigaku Ultima III software package was employed. The polarization resistance (Tafel curves) and electrochemical impedance spectra (EIS) in a saline (3.5% NaCl solution) environment were obtained in terms of open-circuit potentials. The Tafel curves were obtained using a PG TEXCORR 4.1 Potentiostat-Galvanostat system at room temperature with a cell composed of the working electrode, which had an exposed area of 1 cm². The curves were obtained using a Ag/AgCl reference electrode and a platinum wire as a counter-electrode



Fig. 1. X- Ray diffraction patterns of the CrN coatings grown on AISI 304 substrates at 0.4 Pa while varying the $Ar-N_2$ gas mixture.

with a scanning step of 1 mV/s. The Tafel diagrams were obtained using a scan speed of 0.126 mV/s over a voltage range between -0.25 and 1 with an exposure area of 1 cm^2 in an electrochemical solution of 3.5% p/p of NaCl.

3. Results and discussion

3.1. XRD analysis

Fig. 1 presents the X ray diffraction patterns of the CrN coatings produced on AISI 304 stainless steel at 4 Pa for different mixtures of $Ar-N_2$. Diffraction peaks corresponding to the (111), (200), (220), (311) and (222) planes for the cubic CrN structure were identified. These orientations are based on the JCPDF 00-011-0065 file for the CrN ICDD cards. These results are consistent with studies performed by Hones et al. [22]. The diffraction patterns reveal a preferential orientation in the (111) direction, which is characteristic of the fcc-CrN phase [23,24]. The identification of the peaks was performed using the ICSD crystal structure database [25]. A more detailed analysis of the crystallographic orientation of the α -CrN phase is presented in Fig. 2 (a)-(d), in which the peaks that correspond to the (111), (200), (220) and (311) planes for nitrogen flows of 1.5, 2 and 5 sccm are magnified. Based on the peaks with the highest intensities (Fig. 2(a) and (c)), it can be concluded that the peaks tend to shift toward lower diffraction angles as the flow of nitrogen increases. This behavior is difficult to observe in the other peaks (Fig. 2(b) and (d)) because of their low intensity and the noise present in the diffraction patterns. This tendency is attributed to the insertion of nitrogen into the CrN structure, which produces an increase in the interplanar distance and in the lattice parameter. Subsequently, an increase in the nitrogen concentration generates an increase in the movement of the surface atoms on the substrate. This effect increases the energetic state of the atoms, which generates a structural distribution where there is a lower Gibbs energy. This distribution produces an increase in the density and in the interatomic distance [26] that causes a compressive stress; next, at low values of nitrogen flux, coatings with less intrinsic stress were Download English Version:

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