



Contents lists available at ScienceDirect

Surface & Coatings Technology

journal homepage: www.elsevier.com/locate/surfcoat

Investigation of the photocathodic protection of a transparent ZnO coating on an AISI type 304 stainless steel in a 3% NaCl solution

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ARTICLE INFO

Article history:

Received 15 August 2016

Revised 16 January 2017

Accepted in revised form 24 January 2017

Available online xxxx

Keywords:

Photoelectrochemistry

Corrosion

Thin film

Deposition

Spray pyrolysis

Nanostructure

ABSTRACT

The coating of a suitable n-type oxide semiconductor on a metal substrate can provide a photocathodic protection for which the coating protects the metal as a non-sacrificial photoanode under light illumination. In this work, the photocathodic protective characteristics of a transparent ZnO coating on an AISI type 304 stainless steel (SS304), in a 3% NaCl solution, was investigated. The ZnO film was coated on a SS304 substrate by spray pyrolysis using a precursor solution of zinc acetate dihydrate and methanol. The substrate temperatures were varied at 200 and 400 °C during the coating processes. The ZnO coating at 200 °C (200ZnO) exhibited dense nano-powdery films with non-crystalline structures, while the ZnO coating at 400 °C (400ZnO) showed loosely-packed needle-like nano-granular deposits with polycrystalline structures. The photocathodic characteristics of the coatings were analyzed by the potentiodynamic linear polarization and open circuit potential (OCP) measurements under dark and UV illumination. Both 200ZnO and 400ZnO coatings exhibited photocathodic characteristics with lower corrosion potentials and higher current densities under UV illumination. In the anodic polarization region, 200ZnO coatings exhibited passivity similar to bare SS304. Yet, the microstructure of 200ZnO coatings after the polarization test showed traces of partial dissolution of ZnO. 400ZnO provided no passivity and poor corrosion resistance as the coating completely dissolved and the exposed SS304 was uniformly corroded under both dark and UV conditions.

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1. Introduction

Stainless steels are a class of steels that are widely used under corrosive environment due to their good corrosion resistance in various corrosive environments. However, in the presence of Cl⁻ ions, they are susceptible to localized corrosion, e.g. pitting corrosion. As a result, ceramic or organic coatings are widely applied to protect stainless steels under such environment. Recently, the photocathodic protection by the coating of an n-type oxide semiconductor on a metal substrate has gained a lot of attentions due to its ability to protect the coated metal as a non-sacrificial photoanode under light illumination without using sacrificial anode materials or external electric power [1–9].

The photocathodic protection was first reported by Yuan and Tsujikawa [10] with the corrosion prevention of Cu by an n-type TiO₂ coating under UV illumination. Under illumination, electron-hole pairs

can be generated in the TiO₂ coating (with bandgap energy of 3.2 eV). The holes can oxidize H₂O in the electrolyte to form O₂ and H⁺:



The electrons can migrate toward the metal substrate, whose Fermi energy (E_F) must be lower than TiO₂, to shift its potential lower than the potential at which the metal begins to oxidize; as a result, the metal substrate becomes more cathodic and stable. The electron can also react with H⁺ or H₂O to form H₂ or OH⁻:



As a result, the TiO₂ coating exhibits a non-sacrificial photoanodic characteristic as the anodic reaction occurs via the oxidation reaction of H₂O with photogenerated holes, without the dissolutions of either TiO₂ or the metal substrate [10]. For the photocathodic protection to become effective, the open circuit potential (OCP) of the metal substrate must be higher than that of the TiO₂ coating (E_F of the metal must be

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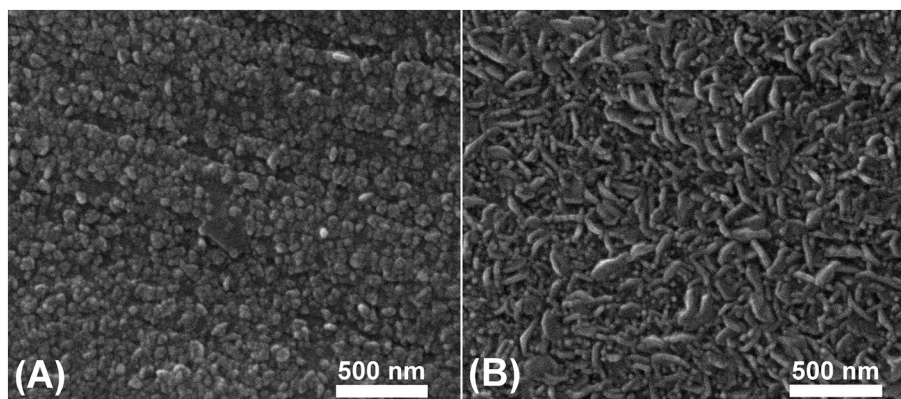


Fig. 1. SEM microstructures of ZnO coatings on SS304: (a) as-deposited coating at 200 °C, (b) as-deposited coating at 400 °C.

lower than that of TiO_2) to ensure electron migration to the metal. As a result, the photocathodic protection of TiO_2 coatings can potentially be effective for copper, nickel, or stainless steel, especially under chloride environment.

The photocathodic protection can be applied either by a direct coating [2–4,11,12] or as a coupling cell [6,13–15]. For the coupling cell method, the photocathodic film is placed in a separated photoelectrochemical cell that is directly exposed to UV or visible light, while the corrosion cell that contains a metal working electrode does not need to be illuminated. The photocathodic film is electrically coupled with the metal working electrode for electron transfer. The coating of a TiO_2 film on a stainless steel or copper is commonly carried out by a sol-gel technique [3,11,12]. The TiO_2 film can also be fabricated on a Ti substrate by the anodization technique [5,7–9,14,16–18]. The resultant TiO_2 films normally require post heat treatment at temperature above 450 °C to form anatase phase that exhibits better photocatalytic properties than other TiO_2 isomorphs [2]. The optimal annealing temperature range to maximize the anatase content was reported to be 450–500 °C [3].

ZnO can be an alternative n-type semiconductor material with low cost, low toxicity, proper bandgap energy of 3.37 eV, and good photocatalytic activity [19,20]. Sun et al. [21] reported that a powdery ZnO electrode in a coupling cell of a 3.5% NaCl solution under light illumination shifted its OCP to be more negative than that of SS304. Therefore, the photogenerated electrons could transfer to SS304 and provide photocathodic protection for SS304. The powdery ZnO electrode was produced by a co-precipitation method and subsequently annealed at 800 °C for 3 h [21]. Xu et al. [15] reported that pure ZnO and hybrid TiO_2/ZnO coatings on SS304 substrates decreased the corrosion potentials by -0.16 and -0.42 V, respectively. The TiO_2/ZnO composite film on SS304 was reported to increase light absorption and reduce electron-hole recombination rate that enhanced the photocathodic protection resulting in better photocathodic protective performance than that of ZnO coating. Both ZnO and TiO_2/ZnO coatings were spin-coated on substrates with sol and subsequently calcined at 500 °C for 2 h [15].

A high temperature coating process can adversely affect the structures and properties of metal substrates. It is well known that, in the temperature range between 500 and 800 °C, chromium carbides may precipitate along the grain boundaries of stainless steels that can affect their mechanical properties and corrosion characteristics. Moreover, high temperature exposure under ambient atmosphere can cause surface oxidation that may result in undesirable surface colors and appearance. ZnO thin films can be deposited at low temperature by spray pyrolysis technique. Spray pyrolysis is a fast, cost effective technique that can be applied on a large scale to produce thin film coatings [22]. ZnO layers were successfully prepared by a spray pyrolysis in air on a substrate temperature of 150 °C using a 0.1 M $\text{Zn}(\text{NO}_3)_2$ aqueous solution as precursor [23]. Studenikin et al. [24] reported that, for the

spray pyrolysis using aqueous zinc nitrate precursors, there was a critical temperature (T_c) of 180 °C below which the thermal decomposition to ZnO was incomplete, while films grown above T_c showed ZnO polycrystalline structures.

To date, there is no report on the photocathodic characteristics of a direct ZnO coating on a stainless steel substrate by spray pyrolysis at low temperature. In this work, we investigated the photocathodic characteristics of ZnO coatings on a SS304 substrate in 3% NaCl solution under dark and UV illumination. The coatings were deposited on SS304 substrates by spray pyrolysis at 200 and 400 °C. Their photocathodic characteristics were investigated and compared along with their morphological and phase analyses.

2. Material and methods

The ZnO thin film was coated on a 2×2 cm AISI type 304 stainless steel (SS304) by a pneumatic spray pyrolysis technique. For the spray pyrolysis system, a precursor solution of 0.05 M zinc acetate dihydrate in methanol solvent was fed by a syringe pump at 4 ml/min, to mix with a carrier gas at a spray nozzle forming micro-droplet sprays of the precursor solution. The carrier gas, with a flow rate of 0.2 kg/cm², was directly compressed from the atmosphere using air filters to remove moisture and dust contamination. The precursor solution was sprayed on a heated SS304 substrate for 10 min to coat ZnO thin films on the substrate. The ZnO coatings were prepared at varying substrate

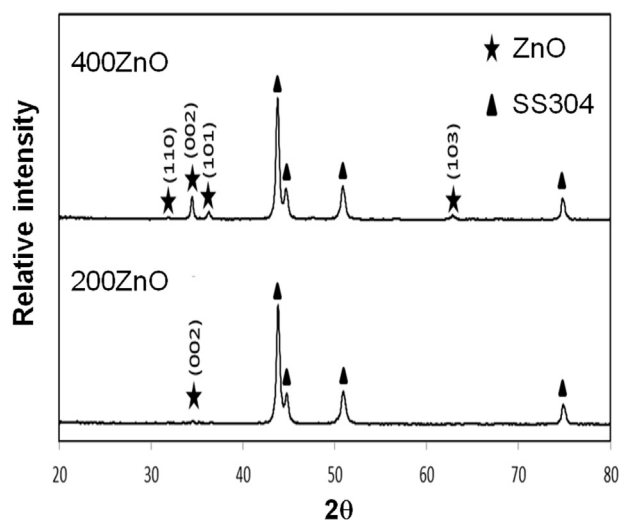


Fig. 2. Representative XRD patterns of the ZnO coatings at 200 °C (200ZnO) and 400 °C (400ZnO).

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