



# Effect of sodium benzoate on corrosion behavior of 6061 Al alloy processed by plasma electrolytic oxidation



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## ARTICLE INFO

### Article history:

Received 21 August 2015

Revised 28 October 2015

Accepted in revised form 3 November 2015

Available online 9 November 2015

### Keywords:

6061 Al alloy

Plasma electrolytic oxidation

Sodium benzoate

Corrosion

## ABSTRACT

The effect of sodium benzoate (NaBz) in an electrolyte on the corrosion behavior of a 6061 Al alloy coated by plasma electrolytic oxidation (PEO) was investigated. For this purpose, a PEO coating was performed on the present sample under alternating current conditions in an alkaline–aluminate electrolyte with and without NaBz. Regardless of the addition of NaBz, the microstructural observation showed that the coating layers consisted of outer and inner layers. When the alloy was coated in the electrolyte with NaBz, a higher fraction of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> was found in the coating film as compared to the sample coated in the electrolyte without NaBz. In addition, the coating of the sample processed in the electrolyte with NaBz became dense, which resulted in better corrosion resistance than the coating without NaBz. The potentiodynamic polarization test showed that the coating layer formed on the electrolyte with NaBz exhibited superior corrosion resistance to that without NaBz. The corrosion behavior of the PEO-coated sample could be explained using the equivalent circuit model.

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## 1. Introduction

Al alloys have been used widely in the aircraft industry owing to their high strength and low density [1,2]. On the other hand, their limited surface hardness and relatively low corrosion resistance have restricted their practical applications. Plasma electrolytic oxidation (PEO) is a novel electrochemical surface treatment process which was used widely to produce ceramic coatings on Al alloys to overcome those limitations. The structure of the ceramic coatings produced by PEO process is affected by the various processing conditions, including chemical composition and concentration of the electrolyte, substrate, coating time, and electrical parameters, such as current density, current mode, and current frequency [3–10]. Among them, the chemical composition of the electrolytes plays an important role in changing the morphology and electrochemical properties of the ceramic coating produced by PEO. Wang et al. [11] reported that the corrosion resistance of LY12 Al alloy processed by PEO was improved by the addition of NaF to the sodium aluminate (NaAlO<sub>2</sub>) electrolyte. Gang et al. [12] reported that NaAlO<sub>2</sub> in the electrolyte could be responsible for the formation of high amount of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> on Al alloy processed by PEO. Liang et al. [13] and Guo et al. [14] examined the effects of AlO<sub>2</sub><sup>-</sup> in the electrolyte on the electrochemical properties of PEO coatings, and reported that the addition of AlO<sub>2</sub><sup>-</sup> caused the formation of the spinel MgAl<sub>2</sub>O<sub>4</sub> which improved the corrosion resistance of PEO coatings. On the other hand, a variety of organic compounds and other special chemical additives have been used to modify the electrolyte solution in order to impart desirable qualities during PEO coatings,

particularly better corrosion resistance [15]. Wu et al. [16] reported that the addition of glycerol to the silicate electrolyte led to excellent corrosion resistance of AZ91 Mg alloy via PEO coating. Bai and Chen [17] examined the effect of hexamethylenetetramine as additive on the structure and corrosion properties of AZ91 Mg alloy processed by PEO in the alkaline silicate phosphate electrolyte. They reported that hexamethylenetetramine was a useful additive to eliminate cracks and reduce the size of micro pores in coatings, giving rise to better corrosion protection.

Liu et al. [18] reported that the addition of sodium benzoate (NaBz) to an alkaline–borate electrolyte would lead to a significant improvement in the corrosion resistance of the coatings formed on AZ91 Mg alloy via PEO process. This was attributed to the fact that both pore size and surface roughness decreased with the addition of NaBz. However, little information regarding the effect of NaBz addition in an alkaline–aluminate electrolyte on the microstructure and electrochemical response of the coating layers of Al alloys subjected to PEO was available. Therefore, the objective of this study was to fabricate Al<sub>2</sub>O<sub>3</sub> coatings formed on 6061 Al alloy via PEO process in the alkaline–aluminate electrolyte containing NaBz. The characteristics of the present coatings were analyzed in relation to structure, composition, and corrosion assessment.

## 2. Experimental procedures

Prior to PEO coating, 6061 Al alloy samples with dimensions of 30 mm × 20 mm × 4 mm were polished mechanically with #1200 SiC paper, rinsed with distilled water, and cleaned ultrasonically in pure ethanol. A series of PEO coatings were carried out under AC condition at a frequency of 60 Hz. The present electrolytes used in this study

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comprised  $\text{NaAlO}_2$  and  $\text{KOH}$  as a base electrolyte with and without the addition of  $\text{NaBz}$  as shown in Table 1. Here, the conditions of the electrolytes without and with  $\text{NaBz}$  corresponded to Baths A and B, respectively. For both electrolytes, the values of pH and electrical conductivity were approximately 13 and 24 mS/cm, respectively. The temperature of the electrolyte was held at 283 K to stabilize the electrochemical reactions during PEO process. The applied current density was fixed to  $100 \text{ mA/cm}^2$  for 180 s. After PEO treatment, the microstructures of the surfaces and cross-section images were examined by scanning electron microscopy (SEM, HITACHI, S-4100). The thickness of the oxide layer was measured from the cross-section morphologies. The program of Image Analyzer 1.33 was used to measure and calculate the pore size and proportion on the surface of the coatings. A non-contact 3-D surface measurement system (U-surf, NANOFOCUS) was used to measure the surface roughness of the PEO-coated samples. The surface roughness was characterized commonly by the arithmetic root mean square roughness ( $R_q$ ). The phase structure was analyzed by X-ray diffraction (XRD, RIGAKU, D-MAX 2500) with a step size of  $0.05^\circ$  and a scan range of  $20^\circ$  to  $90^\circ$ . The chemical composition of the oxide layer was examined by X-ray photoelectron spectroscopy (XPS, VG Microtech, ESCA 2000). The electrochemical measurements were evaluated in a 3.5 wt.%  $\text{NaCl}$  solution at pH of 7 utilizing three different electrodes: a coated sample with an exposed area of  $1 \text{ cm}^2$  as the working electrode, a platinum plate as the counter electrode, and an  $\text{Ag/AgCl}$  electrode as the reference electrode. The general corrosion behavior of the coating was evaluated by potentiodynamic polarization and electrochemical impedance tests (Gamry Instruments). The polarization curves were measured from  $-0.25$  to  $0.4 \text{ V}$  with respect to the open circuit potential at a scan rate of  $1 \text{ mV/s}$ . Electrochemical impedance spectroscopy (EIS) was conducted from  $10^6$  to  $0.1 \text{ Hz}$  at an interval of 10 points/decade with a  $10 \text{ mV rms}$ .

### 3. Results and discussion

#### 3.1. Voltage-time curve

Fig. 1 shows the voltage as a function of the coating time of the samples coated in Baths A and B. The coating voltages increased rapidly up to  $\sim 225 \text{ V}$  due to the rapid electrochemical formation of an initial insulating oxide layer, obeying Ohm's rule [19]. The weak parts of the oxide layer would be broken down after the applied voltage exceeded the breakdown voltage, resulting in the occurrence of a spark discharge phenomenon. For Baths A and B, the breakdown voltages were similar to each other ( $\sim 225 \text{ V}$ ) owing to the reasonably similar values of the electrical conductivity [4]. The time to reach the breakdown voltage could reflect the ease of the dielectric barrier layer to form and plasma discharge to take place [20]. Therefore, the breakdown phenomena in Bath A occurred earlier than in Bath B despite the similar breakdown and final voltages of two conditions. The reason for the present delay in Bath B might be presumably attributed to the fact that the high dissolubility of Al alloy in the benzoates would prevail over the oxidation reaction [21].

#### 3.2. Coating morphology

Fig. 2 displays the surface morphologies of the oxide layers produced by PEO in Baths A and B. Regardless of the addition of  $\text{NaBz}$ , the PEO-coated samples showed a number of micro pores which were typical

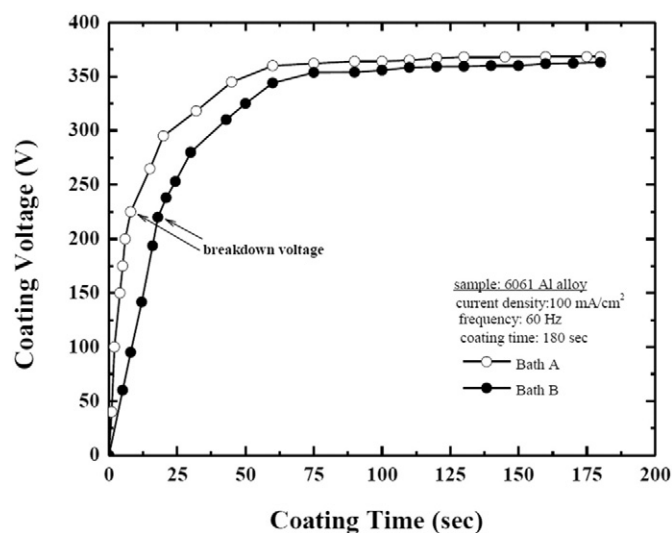
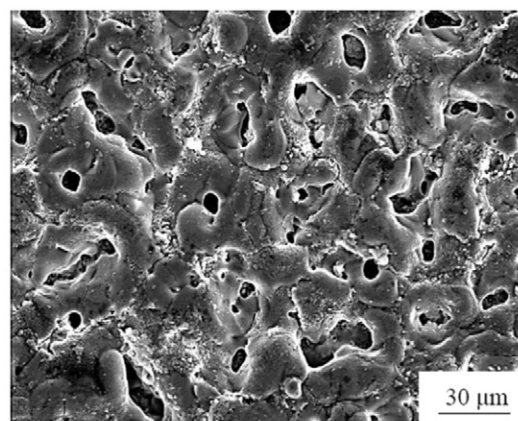
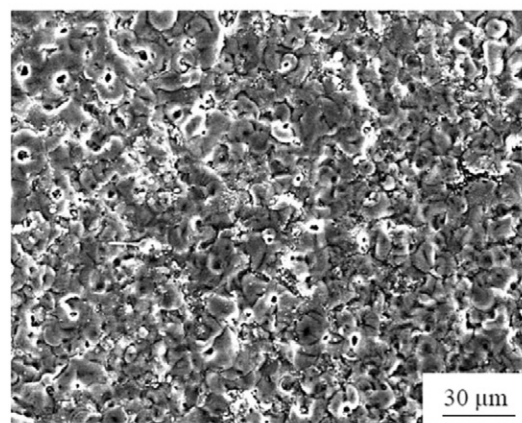


Fig. 1. Voltage-time curves of 6061 Al samples during PEO coatings in Baths A and B under AC condition for 180 s at a frequency of 60 Hz.

for the samples after PEO coating [3–5]. The micro pores would be originated from the molten oxide and gas bubbles thrown out of the discharge channels. Some micro cracks also appeared on the surface of the samples treated in Bath A, which were attributed to the significant



(a)



(b)

Fig. 2. Surface morphologies of the oxide layer of 6061 Al samples by PEO coatings in (a) Bath A and (b) Bath B.

Table 1

Chemical compositions of the electrolytes used for the present PEO coatings.

Electrolyte	Composition/(g/L)		
	$\text{NaAlO}_2$	$\text{KOH}$	$\text{NaBz}$
Bath A	4.098	5.61	0
Bath B	4.098	5.61	1

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