# Effect of Al<sub>2</sub>O<sub>3</sub> Particle Concentration on the Characteristics of Microarc Oxidation Coatings Formed on Pure Titanium

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Microarc oxidation coatings were successfully fabricated on pure titanium substrates by microarc oxidation (MAO) in electrolytes with Al<sub>2</sub>O<sub>3</sub> particles of varying concentrations, which ranged from 0 to 4 g·L<sup>-1</sup> (0, 1, 2, 3, 4 g·L<sup>-1</sup>). The voltage versus time curves, surface and cross-sectional morphologies, thicknesses, microhardnesses, phase compositions, coating adhesive strengths, thermal shock resistances and polarization curves of MAO coatings were investigated. The results revealed that the voltage, the microhardness and the thickness of the coatings increased with the addition of Al<sub>2</sub>O<sub>3</sub> particles. Both the number and size of sintered discs on the coating surface increased gradually with the ascending concentration of Al<sub>2</sub>O<sub>3</sub> particles. The XRD results indicated that the MAO coatings mainly consisted of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and rutile. The corrosion resistance of the coatings was improved, while the addition of Al<sub>2</sub>O<sub>3</sub> particles had less effect on the adhesive strength of the MAO coatings. The study revealed that an appropriate addition of Al<sub>2</sub>O<sub>3</sub> particles in electrolytes is beneficial to promote the performance of coatings.

Keywords: Microarc oxidation; Pure titanium; Al<sub>2</sub>O<sub>3</sub> particles; Performance

# **1. INTRODUCTION**

Titanium and its alloys, due to their high melting points, good corrosion resistance, high strength-to-weight ratios, suitable mechanical properties and good biocompatibilities, are commonly employed in fields such as the marine, aerospace, chemical and biological medicine industries[1-2]. The outstanding corrosion resistance of titanium and its alloys is ascribed to the thin oxide surface layer (1.5-10 nm thick) that is formed naturally on the surface of titanium metal when it is exposed to air at room temperature[3]. However, the thin layer can easily be damaged to failure by exogenic action; thus, these layers hardly meet the performance requirements of service. Furthermore, the safe

working temperature of pure titanium is generally below 300°C[4], and thus its high-temperature performance is inferior, which restricts titanium alloys from even wider application. Microarc oxidation (MAO), also referred to as plasma electrolytic oxidation (PEO), has been considered one of the most cost-saving and environmentally friendly technology to fabricate MAO coatings on the surface of valve metals, such as aluminum, magnesium and titanium, as well as their respective alloys[5-8]. The properties of MAO coatings are controlled by electrolytes, process parameters (such as current density, voltage, pulse frequency and duty cycle), substrate materials, etc.[9-11]. Different additives are added to base electrolytes to dope MAO coatings, which has been identified as an excellent method to modulate the properties of MAO coatings[12-13]. In recent reports, it was found that SiC[14], ZrO<sub>2</sub>[15] and TiO<sub>2</sub>[16] additives were incorporated in the formation of MAO coatings. There are already a few studies examining the effect of Al<sub>2</sub>O<sub>3</sub> particle addition on MAO coatings. Yan[17] found that the corrosion resistance of coatings formed on AZ91D Mg alloy improved with the addition of Al<sub>2</sub>O<sub>3</sub> micropowder, and Al<sub>2</sub>O<sub>3</sub> particles were beneficial to the fast formation of the oxide layer. Ping[18] noted that increasing the Al<sub>2</sub>O<sub>3</sub> micropowder concentration reduced the size of micropores on 6061 aluminum alloy coating surface, and the coating surface became smooth. However, the influence of doping Al<sub>2</sub>O<sub>3</sub> particles on TA2 pure titanium was never reported.

In this investigation, MAO coatings were fabricated on pure titanium substrates at different  $Al_2O_3$  particle concentrations in  $Na_2SiO_3$ -NaOH electrolyte system. In addition, the effect of  $Al_2O_3$  particle concentration in the electrolytes on the voltage, surface and cross-sectional morphologies, surface microhardness, coating thickness, phase composition, coating adhesive strength, thermal shock resistance and corrosion resistance of MAO coatings on pure titanium materials was investigated, and the doping mechanism of  $Al_2O_3$  particles was also discussed.

#### 2. MATERIALS AND METHODS

### 2.1. Preparation of MAO coating

Rectangular samples of TA2 pure titanium (with chemical composition by wt%: Fe $\leq$ 0.30%, C $\leq$ 0.10%, O $\leq$ 0.25%, N $\leq$ 0.05%, H $\leq$ 0.015% and Ti balance) were used as substrate materials in the experiments. The specimens, with dimensions of 15×15×2 mm, were polished and degreased before the MAO process. The base electrolyte consisted of 18 g·L<sup>-1</sup> Na<sub>2</sub>SiO<sub>3</sub>, 1 g·L<sup>-1</sup> NaOH and 3 ml·L<sup>-1</sup> C<sub>3</sub>H<sub>8</sub>O<sub>3</sub>. The resin was used to mount the samples to observe cross-sectional morphologies. The concentration of Al<sub>2</sub>O<sub>3</sub> particles in the base electrolyte was 0 g·L<sup>-1</sup>, 1 g·L<sup>-1</sup>, 2 g·L<sup>-1</sup>, 3 g·L<sup>-1</sup>, and 4 g·L<sup>-1</sup>. In addition, the Al<sub>2</sub>O<sub>3</sub> particles with an approximate size of 10-40 µm were stirred continuously during the experiment in order to keep uniform dispersal in the electrolyte. The MAO process was carried out using a pulsed electrical power that provided a positive pulse voltage. The MAO coatings were fabricated under a peak current density of 5 A·dm<sup>-2</sup> for 30 min, a fixed frequency of 100 Hz and a duty cycle of 40%. The temperature of the electrolyte was kept below 30°C by a heat exchange system during the MAO process. After MAO treatment, the specimens were rinsed with hot water at 95°C for 20 min and then dried in warm air.

#### 2.2. Coating analysis

The surface and cross-sectional morphologies of MAO coatings in the electrolytes with different concentration of  $Al_2O_3$  particles were observed by a Scanning Electron Microscope (SEM, ZEISS EVO MA15) equipped with an Energy Dispersive Spectrometer (EDS, OXFORD 20). The phase composition of the coatings was investigated by X-ray Diffraction (XRD, DX-2700B). Diffraction data were acquired over scattering angle 20 from 10° to 80°, and the scanning speed was  $0.05^{\circ} \cdot s^{-1}$ . Scratch tests, which could evaluate the adhesion between coatings and substrate, were implemented by multifunctional surface performance tester (MFT-4000) with automatic loading from 0-20 N at a loading rate of 10 N·min<sup>-1</sup>. In addition, thermal shock tests of the coatings were investigated by using a chamber electric furnace (SX-10-12), which was heated to 300°C for 10 min and then cooled down in 25°C water for 50 cycles. The microhardness and thickness of the coatings were detected by digital microhardness tester (HVS-1000) at 1 N for 15 s and a Digital Thickness Gauges (TT230), respectively. Furthermore, an Electrochemical Workstation (IM6, Netherlands) was used to acquire polarization curves in 3.5% NaCl solution at room temperature. The polarization curve scanning rate was 1.5 mV·s<sup>-1</sup> with a potential range from -1.0 V to -0.4 V.

#### **3. RESULTS AND DISCUSSION**

#### 3.1. Voltage-time response



**Figure 1.** Voltage-time curves at different concentrations of Al<sub>2</sub>O<sub>3</sub> particles: (a) original drawing, (b) enlarged drawing

Fig. 1 shows the corresponding voltage response of different  $Al_2O_3$  particles concentrations during the MAO process. It can be observed that the voltage increased gradually with the increase of  $Al_2O_3$  particles concentration, and the maximal voltage was approximately 430V at the  $Al_2O_3$  particles concentration of 4 g·L<sup>-1</sup>. At the initial anodic oxidation and spark oxidation stage, the voltage increased linearly to accomplish dielectric breakdown and electric arc discharge and then quickly evolved to a microarc oxidation stage in which the voltage tended to be smooth and steady. The surface charge of  $Al_2O_3$  particles in the electrolytes was negative[18], which easily led to the movement of  $Al_2O_3$  particles towards the anode, and the  $Al_2O_3$  particles were adsorbed gradually on the surface of the specimen through the combined effects of the electric field and mechanical stirring. The  $Al_2O_3$  particles that were adsorbed on the coating surface heightened the resistance of the coatings, which caused the voltage of the coatings to consequently increase in accordance with Ohm's Law.

#### 3.2. The surface and cross-sectional morphologies of coatings

Fig. 2 represents the surface morphologies and major element content of MAO coatings at different Al<sub>2</sub>O<sub>3</sub> particles concentrations. The coating surface was relatively smooth, and it was accompanied by small holes when no  $Al_2O_3$  particles were added, as shown in Fig. 2(a). While the size and number of sintered discs increased gradually with the ascending concentration of Al<sub>2</sub>O<sub>3</sub> particles, as shown in Fig. 2(b)-(e), the number of holes decreased. The increased number of sintered discs on the surface of the MAO coatings with adding Al<sub>2</sub>O<sub>3</sub> particles was ascribed to the rise of voltage, which led to higher energies in individual discharge channels during MAO processing. Yan[17] also found that, after adding Al<sub>2</sub>O<sub>3</sub> particles, the hole size and the roughness of coatings on AZ91D Mg alloys increased due to the rising energy. The temperature of micro areas boosted to  $10^3$ - $10^4$  K[19] during the spark discharge process, which was sufficient to melt both the micro areas of the coatings and the adsorbed Al<sub>2</sub>O<sub>3</sub> particles. The melted Al<sub>2</sub>O<sub>3</sub> particles filled up the discharge channels, which reduced the number of holes and enhanced the hardness of the coatings. With the further addition of Al<sub>2</sub>O<sub>3</sub> particles, more energy was obtained owing to the anabatic voltage, which indicated that discharge breakdown was more likely to come up, and then the substrate was ejected easily through the discharge channels to form more sintered discs. As a consequence, the number of sintered discs increased and the number of holes decreased. Fig. 2(a)-(e) also exhibits the major elemental content of the MAO coatings. It is obvious that the content of Al and Si increased, the content of O had very little change, and the content of Ti reduced gradually with the addition of Al<sub>2</sub>O<sub>3</sub> particles. Apparently, the Si and Al were from the electrolyte solution, which demonstrated that Al<sub>2</sub>O<sub>3</sub> particles were involved in the formation of the MAO coatings; this influenced the reaction between the electric arcs and the substrate, which gave rise to the change of elemental content in the coatings. In addition, the voltage increased with the increase of Al<sub>2</sub>O<sub>3</sub> particles, and the thickness of the coatings grew gradually. Thus, the elements in the electrolyte solution were easier to enter into the coatings, whereas it was more difficult for the substrate Ti to transmit to the coating surface owing to the thickening of coatings. Therefore, the content of Al and Si showed a relatively upward trend, but the content of Ti ultimately decreased with the addition of Al<sub>2</sub>O<sub>3</sub> particles. The possible reactions during MAO process are as follows[18, 20]:

$$\begin{array}{ll} H_{2}O \rightarrow H^{+} + OH^{-} & (1) \\ 2OH^{-} \rightarrow O^{2^{-}} + H_{2}O & (2) \\ Ti \rightarrow Ti^{4^{+}} + 4e^{-} & (3) \\ Ti^{4^{+}} + 2OH^{-} + 2H_{2}O \rightarrow TiO_{2} + 2H_{3}O^{+} & (4) \\ Ti^{4^{+}} + 2O^{2^{-}} \rightarrow TiO_{2} & (5) \end{array}$$

$$SiO_3^{2-} + Al_2O_3 + H_2O \rightarrow H_2SiO_3 + 2AlO_2^{-}$$
(6)  
$$H_2SiO_3 \rightarrow H_2O + SiO_2$$
(7)



**Figure 2.** Surface morphologies and element content of MAO coatings with different concentration of Al<sub>2</sub>O<sub>3</sub> particles: (a)  $0 \text{ g} \cdot \text{L}^{-1}$ , (b)  $1 \text{ g} \cdot \text{L}^{-1}$ , (c)  $2 \text{ g} \cdot \text{L}^{-1}$ , (d)  $3 \text{ g} \cdot \text{L}^{-1}$  and (e)  $4 \text{ g} \cdot \text{L}^{-1}$ .

Cross-sectional microstructures of MAO coatings with different concentrations of  $Al_2O_3$  particles were prepared by resin, as shown in Fig. 3, and the oxidation coating was embedded between the resin and the pure titanium substrate. It is clear that the thickness of the MAO coatings demonstrated a rising tendency with ascending concentrations of  $Al_2O_3$  particles. More energy was acquired with the increasing voltage, thus the growth rate of the MAO coatings was expedited, which resulted in the increase of coating thickness in accordance with the following thickness analysis.

Although there were some cracks caused by thermal shrinkage of the resin between the coatings and the resin, the combination between the coatings and the substrate was relatively compact.





**Figure 3.** Cross-sectional images and Al element distribution of MAO coatings with different concentrations of Al<sub>2</sub>O<sub>3</sub> particles: (a)  $0 \text{ g} \cdot \text{L}^{-1}$ , (b)  $1 \text{ g} \cdot \text{L}^{-1}$ , (c)  $2 \text{ g} \cdot \text{L}^{-1}$ , (d)  $3 \text{ g} \cdot \text{L}^{-1}$  and (e)  $4 \text{ g} \cdot \text{L}^{-1}$ .

3.3. The microhardness and thickness of coatings



**Figure 4.** The microhardness and thickness of MAO coatings prepared with different Al<sub>2</sub>O<sub>3</sub> particles concentrations

Microhardness and thickness tests were carried out on specimens prepared at different Al<sub>2</sub>O<sub>3</sub> particles concentrations, which are depicted in Fig. 4. The substrate hardness of pure titanium is only 160.1 HV, while the hardness of MAO coating without Al<sub>2</sub>O<sub>3</sub> particles is approximately 228.4 HV. The hardness of all the MAO coatings with Al<sub>2</sub>O<sub>3</sub> particles is much higher than that of the MAO coating without Al<sub>2</sub>O<sub>3</sub> particles. Meanwhile, with the addition of Al<sub>2</sub>O<sub>3</sub> particles, the average microhardness linearly increased, which was greatly in accord with Yan's research results[17]. The MAO coating with 4 g·L<sup>-1</sup> Al<sub>2</sub>O<sub>3</sub> particles exhibited the highest hardness of 392.1 HV, which was more than twice as hard as the substrate. The ascending of microhardness is mainly ascribed to the significant increase of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, which exhibits higher hardness than pure titanium and titanium dioxide. On the other hand, the thickness of coatings increased with the rising of Al<sub>2</sub>O<sub>3</sub> particles concentration, in accordance with the trend of the voltage. At the local high temperature, the melted Al<sub>2</sub>O<sub>3</sub> particles were incorporated into the MAO coating through discharge channels and accelerated

the growth of the coating, which led to the increase of coating thickness with ascending  $Al_2O_3$  particles concentration.

# 3.4. The phase composition of coatings

Fig. 5 illustrates the phase patterns of the MAO coatings prepared in electrolytes with different concentration of Al<sub>2</sub>O<sub>3</sub> particles. The analysis results indicate that the MAO coatings were mainly composed of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and rutile. The peak of Ti coming from the substrate was detected because X-rays can penetrate into the substrate through the MAO coatings[21]. In addition, the peak intensity decreased as the concentration of Al<sub>2</sub>O<sub>3</sub> particles increased, which revealed that the coatings grew thicker; this was in accordance with the results of the coating thickness analysis. When  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> was added to the electrolytes, it gradually transformed to  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> due to the local high temperature of discharge[20], as shown in equation (9). The detection of SiO<sub>2</sub> demonstrated that SiO<sub>3</sub><sup>2-</sup> existed in the electrolyte was involved in the chemical reactions, which we have presented in the chemical equations (6), (7). And the peak intensities of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> elevated slightly with the addition of Al<sub>2</sub>O<sub>3</sub> particles. The rising trend of the two phases was caused by the higher voltage, which demonstrated that higher temperature would be reached, more severe reactions would occur, and an easier transformation between the phases would ensue. In addition, the analysis results were in good accord with the EDS analysis, which illustrated that the content of Si gradually increased owing to the transition from SiO<sub>3</sub><sup>2-</sup> to SiO<sub>2</sub>.



$$\gamma - Al_2 O_3 \xrightarrow{800 - 1200^{\circ}C} \alpha - Al_2 O_3 \tag{9}$$

Figure 5. XRD spectra of the MAO coatings with different Al<sub>2</sub>O<sub>3</sub> particles concentrations

# 3.5. The coating adhesive strength

Fig. 6 shows the coating adhesive strength with the addition of  $Al_2O_3$  particles. It is evident that the addition of  $Al_2O_3$  particles had little effect on the coating adhesive strength of MAO coatings. It

was still found that through the scratch testing, the coating adhesive strength increased slightly with ascending  $Al_2O_3$  particles concentrations. During the microarc oxidation process, the substrate titanium could be directly transformed into crystalline titanium dioxide so that there were no large voids at the interface between the coatings and the substrate[22]; and the lattice matching between the two was also relatively good, so the coatings had a good adhesive strength with the substrate. On the other hand, it has been reported that the coating adhesive strength is related to the variation of the thickness, microhardness and compactness of the coatings[18]. The addition of  $Al_2O_3$  particles provided both thickneed coatings and substrates. Hence, the coatings with the addition of  $Al_2O_3$  particles exhibited better coating adhesive strengths compared to the coatings without the addition of  $Al_2O_3$  particles.



Figure 6. Variation in the coating adhesive strength with different Al<sub>2</sub>O<sub>3</sub> particles concentrations

## 3.6. The thermal shock resistance of coatings

Fig. 7(a) and (b) shows the surface morphologies of the MAO coating at 3  $g \cdot L^{-1} Al_2O_3$  particles under different magnifications after the thermal shock tests. It can be seen that neither cracks nor peeling phenomena occurred on the coating surface of the samples at a temperature of 300°C for 50 cycles, which elaborated that the coatings possessed a good thermal shock resistance. The good thermal shock resistance of the coatings may be attributed to the coating microstructure, as well as the discharge channels and sintered discs (Fig. 2). The sintered discs would absorb thermal shock energy to release the thermal stress of the interface area through the discharge channels, which resulted from different expansion coefficients between the coatings and the substrate[23]. Ying[24] found that thinner coatings yielded better thermal shock resistance. Due to the great difference in hardness between microarc oxidation coatings and substrates, the thicker coatings have larger internal stresses, and thus they generate cracks easily during heating. The thickness of the coating with 3  $g \cdot L^{-1} Al_2O_3$ particles was relatively thin (approximately 10 µm); therefore, the internal stress produced during the thermal shock tests was comparatively small, and the thermal shock resistance of the coating was superior.



**Figure 7.** Surface morphologies of the coating at 3  $g \cdot L^{-1}$  after the thermal shock tests: (a) 200x magnification; (b) 5000x magnification

# 3.7. The corrosion resistance of coatings

Potentiodynamic polarization curves, which are depicted in Fig. 8, were performed in 3.5 wt% NaCl solution to evaluate the corrosion resistance of MAO coatings with different concentrations of  $Al_2O_3$  particles. Table 1 demonstrates the fitting results of the polarization curves by Tafel fitting. According to the Stern-Geary equation[8], the corrosion current density  $i_{corr}$  was automatically calculated as follows:

$$i_{corr} = \frac{\beta_{\alpha} \times \beta_c}{\beta_{\alpha} + \beta_c} \times \frac{1}{R_p}$$
(10)  
Then according to Faraday Law (N–equivalent weight

Then according to Faraday Law (N-equivalent weight,  $\rho$ -density):  $V_{corr}(mil \cdot year^{-1}) = \frac{i_{corr}(A \cdot cm^{-2}) \times N(g) \times 393.7(mil \cdot cm^{-2}) / \rho(g \cdot cm^{-3}) \times 365 \times 24 \times 3600(m \cdot year^{-1})}{96500(C \cdot mol^{-1})}$ (11)  $V_{corr}(mm/a) = \frac{V_{corr}(mil \cdot year^{-1})}{39.37(mil \cdot mm^{-1})}$ (12)

It can be seen that the corrosion rate of microarc oxidation coatings reduced by 2 to 3 orders of magnitude compared with that of the pure titanium substrate, which illustrated that the corrosion resistance was improved dramatically owing to MAO treatment. Furthermore, with the addition of Al<sub>2</sub>O<sub>3</sub> particles, the corrosion rate of the coatings decreased, and the corrosion resistance was thus enhanced. At the same time, Shokouhfar[25] also elaborated that the addition of Al<sub>2</sub>O<sub>3</sub> particles could greatly improve the corrosion resistance of coatings, but the effect was less than that observed foTiO<sub>2</sub>. As the Al<sub>2</sub>O<sub>3</sub> particles concentration increased, the coatings thickened, which impeded the movement of eroding Cl<sup>-</sup> ions to the substrate; this improved the corrosion resistance of the coatings. Simultaneously, as discussed above, the doped  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> gradually transformed into  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> by the higher arc energy resulted from the increase of voltage. In addition, it is well-known that the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> phase has excellent corrosion resistance. Hence, the corrosion resistance of the coatings was improved eventually due to the increase of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> phase.



Figure 7. Polarization curves of MAO coatings with different concentrations of Al<sub>2</sub>O<sub>3</sub> particles

Concentration	$E_{corr}(V)$	$I_{corr}(A \cdot cm^{-2})$	Corrosion rate
$(g \cdot L^{-1})$			$(\mathbf{mm} \cdot \mathbf{a}^{-1})$
Substrate	-0.375	4.496×10 <sup>-7</sup>	$1.564 \times 10^{-2}$
0	-0.274	3.698×10 <sup>-8</sup>	$7.065 \times 10^{-4}$
1	-0.256	7.399×10 <sup>-9</sup>	$5.844 \times 10^{-4}$
2	-0.239	3.529×10 <sup>-8</sup>	4.332×10 <sup>-4</sup>
3	-0.204	$1.694 \times 10^{-8}$	3.649×10 <sup>-4</sup>
4	-0.189	$1.158 \times 10^{-8}$	$2.627 \times 10^{-5}$

Table 1. Results of polarization curve analysis

# 4. CONCLUSIONS

MAO coatings were fabricated on pure titanium substrates by microarc oxidation with different Al<sub>2</sub>O<sub>3</sub> particles concentrations in Na<sub>2</sub>SiO<sub>3</sub>-NaOH electrolyte system. Al<sub>2</sub>O<sub>3</sub> particles were incorporated into MAO coating, which was confirmed by EDS and XRD analysis. In addition, it was evident that to some extent the addition of Al<sub>2</sub>O<sub>3</sub> particles could improve the performance of coatings. The voltage, microhardness and thickness of the coatings increased because the adsorption of negatively charged Al<sub>2</sub>O<sub>3</sub> particles under electrophoresis boosted the electric resistance of the coatings, which facilitated the increase of voltage. Moreover, the increase of electric energy is beneficial to the improvement of the formation rate of coatings. The number of sintered discs increased gradually with the ascending concentration of Al<sub>2</sub>O<sub>3</sub> particles due to higher energy in individual discharge channel. MAO coatings mainly consisted of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and rutile. The corrosion resistance of the coatings was improved owing to the thickening of the coatings and the increase of the anticorrosion phase  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, while the addition of Al<sub>2</sub>O<sub>3</sub> particles had less effect on the coating adhesive strength of MAO coatings.

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### CONFLICTS OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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