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Improvement of surface porosity and properties of alumina films by incorporation of Fe micrograins in micro-arc oxidation

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Abstract

Micro-arc oxidation (MAO) is an effective approach to improve the properties of aluminum and its alloy by forming ceramic films on the surface. However, the oxide layers often have a porous surface structure, which exhibits relatively high friction coefficients. In this work, in order to enhance the surface and mechanical properties of the films produced by micro-arc oxidation, Al_2O_3 coatings embedded with Fe micrograins of different thicknesses were produced on aluminum alloys by adding Fe micrograins into the electrolyte during MAO. Compared to the Al_2O_3 coatings without Fe micrograins, the MAO Al_2O_3 coatings with Fe micrograins are much denser and harder, and the wear resistance is also improved significantly. The enhancement can be attributed to the enhancement of the surface structure and morphology of the MAO Al_2O_3 coatings with embedded Fe micrograins.

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Keywords: Micro-arc oxidation; Al_2O_3 coatings; Fe micrograins; Surface properties

1. Introduction

With increasing demands for lighter and higher specific strength materials, aluminum and its alloys are used in today's industry more widely. However, the poor surface hardness often reduces the lifetime of aluminum components due to wear and fretting damage. Micro-arc oxidation (MAO), a plasma electrolytic surface process, has attracted much interest as an effective technique to improve the wear resistance of aluminum alloys by creating a relatively thick and hard alumina coating on the component surface [1–5]. However, previous studies of MAO aluminum alloys have shown that as-deposited and polished alumina coatings have a relatively high friction coefficient, for example, values of 0.6 and 0.7 against WC/Co and AISI 52100 counter-faces in dry sliding tests, respectively [1]. Therefore, there is a practical need to improve the wear resistance by reducing the friction coefficient because a low friction coefficient is usually required in sliding wear applications. In this respect, deposition of duplex Al_2O_3/DLC coatings on Al alloys for tribological applications using a combined micro-arc oxidation and plasma immersion ion implantation (PIII) technique has been proposed by Nie et al. [6], and duplex surface modification with micro-arc discharge oxidation and magnetron sputtering has also been proposed by Tong et al. [7].

In the MAO process that includes surface micro-arc discharge, diffusion and electrophoresis, various kinds of conventional ceramic coatings containing adventitious elements have been synthesized on metal substrates such as Al, Ti, Mg and their alloys [5,8,9]. Our previous experiments have indicated that some nano-powders such as SiO_2 can be embedded in the porous ceramic coatings on Ti in the form of nanograins by adding SiO_2 nano-powders into the electrolyte. However, the dimensions of the micro-holes are almost the same whether additives are added to the electrolyte or not [10]. In this work, in order to enhance the surface properties and reduce the surface porosity of MAO Al_2O_3 coatings, Fe micrograins were embedded in Al_2O_3 coatings of different thicknesses. The surface and mechanical properties of the MAO Al_2O_3 coatings with and without Fe micrograins were

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characterized, and the enhancement mechanism brought about by Fe micrograin embedding is discussed.

2. Experimental details

Disc samples (20 mm in diameter and 4 mm thick) made of LY2 aluminum alloys consisting of aluminum as the base substrate and approximately 2.6–3.2% Cu, 2.0–2.4% Mg, 0.45–0.7% Mn and <0.8% impurities were ground and polished before MAO. An aqueous solution of electrolytes was prepared with chemically pure $NaWO_3$, $Na_3PO_4 \cdot 12H_2O$ and Fe micrograins (diameters of several μm) at a density of 20 g/l were added to the electrolyte. The treatment was conducted in a stainless steel bath using pulsed and dc power. One end of the power supply output was connected to the bath, and the other one to the samples immersed in the electrolyte. The applied voltage was varied from 300 to 450 V dc. During MAO, a constant average current density of 8 A/dm^2 was maintained on the sample surface by controlling the voltage, and the electrolyte temperature was kept below 50 °C.

The surface morphology of the coatings was characterized by scanning electron microscopy (SEM). X-ray diffraction (XRD) was conducted using the Cu Kα line between 2θ values of 20° and 80° with a step of 0.05°, and X-ray fluorescence spectrometry (XRF) was used to determine the elemental concentrations. The thickness and surface roughness of the coating were measured by an eddy current thickness meter and a Taylor–Hobson/Form Talysurf PGI surface texture tester, respectively. The coating hardness was evaluated by means of Vickers indentation under a load of 200 g. The friction coefficients were measured using a computer-controlled oscillating ball-on-disc scratch tester equipped with a 5 mm WC ball. The wear tests were conducted in air under a load of 4 N with a rotation diameter of 5 mm and sliding speed of 200 rpm.

3. Results and discussion

The XRD spectra acquired from the MAO coatings with and without Fe micrograins are depicted in Fig. 1. These coatings consist mainly of α-Al_2O_3 and γ-Al_2O_3, and the contents vary with the film thickness. The phases are similar in the coatings of

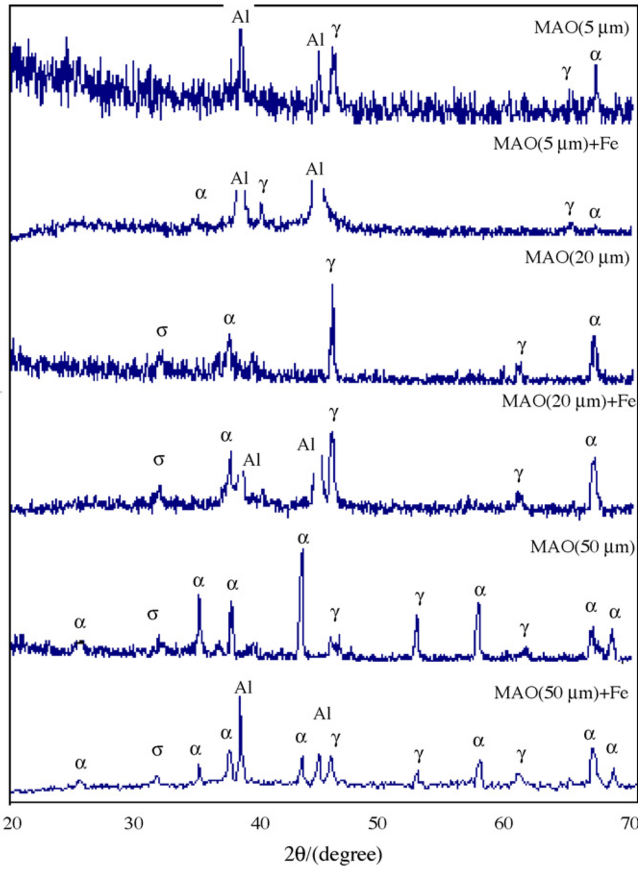

Fig. 1. XRD spectra acquired from the MAO Al_2O_3 coatings of different thicknesses with and without Fe micrograins.

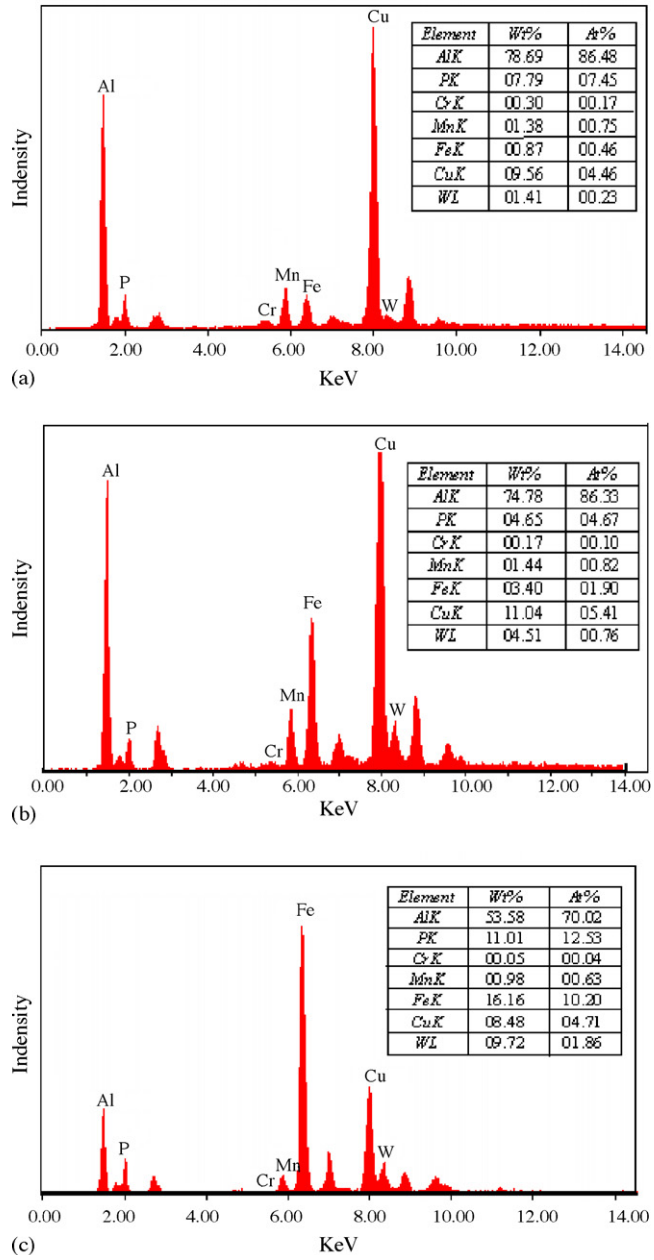

Fig. 2. Elemental concentrations of the Fe-embedded MAO Al_2O_3 coatings of different thicknesses determined by XRF: (a) 5 μm; (b) 20 μm; (c) 50 μm.

the same thickness deposited in the two different solutions. The Fe micrograins are in a Fe-base solid solution state, and no Fe precipitation is detected by XRD. The XRF results shown in Fig. 2 indicate that the Al_2O_3 coatings with Fe micrograins contain large Fe concentrations through diffusion and electrophoresis. The weight percent of Fe increases linearly from 0.87 to 3.4 and 16.16 when the film thicknesses increase from 5 to 20 μm and 50 μm, respectively.

Fig. 3 displays the surface morphology of the MAO Al_2O_3 coatings with and without Fe micrograins obtained by SEM. Crater-like holes of various sizes can be observed in the coatings deposited in the solution without Fe micrograins. Moreover, the thicker the coatings, the larger are these holes. In contrast, the size of these holes in the MAO Al_2O_3 coatings deposited using the solution with Fe micrograins and of similar thicknesses decreases significantly. With increasing coating

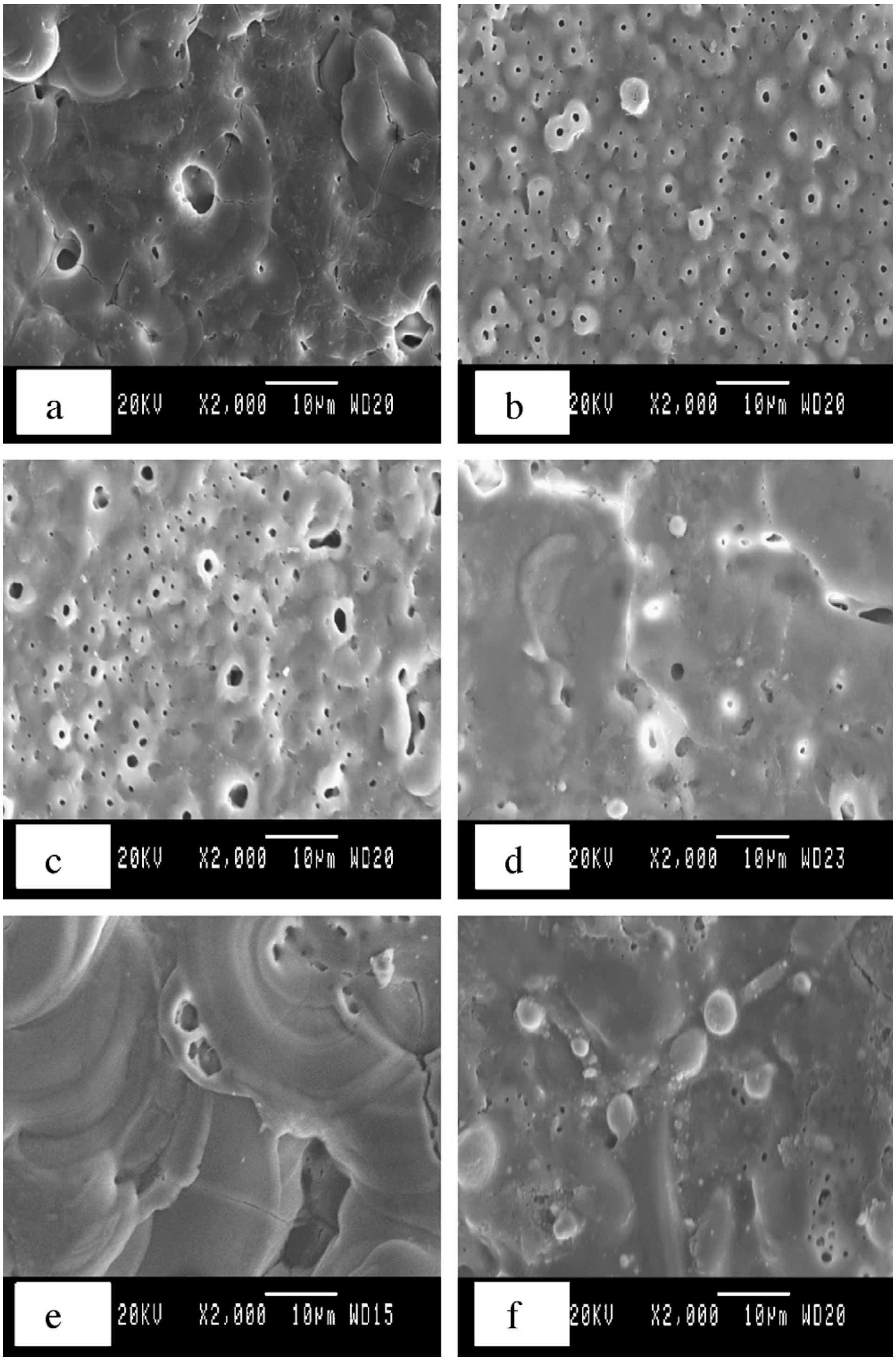

Fig. 3. Surface morphology of the MAO Al_2O_3 coating of different thicknesses with and without Fe micrograins: (a) 5 μm coating without Fe; (b) 5 μm coating with Fe; (c) 20 μm coating without Fe; (d) 20 μm coating with Fe; (e) 50 μm coating without Fe; (f) 50 μm coating with Fe.

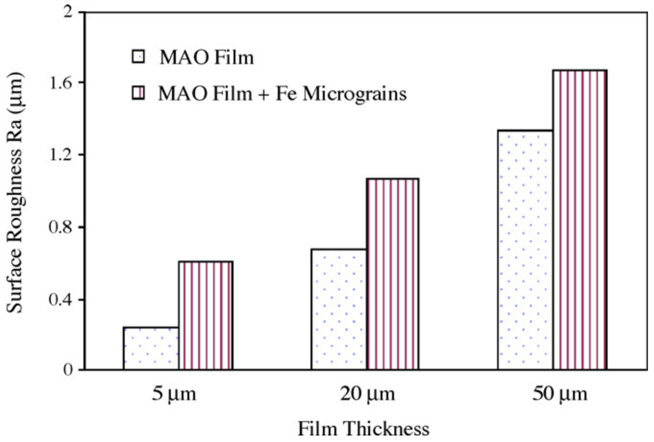

Fig. 4. Relationship between the average surface roughness (R_a) and MAO Al_2O_3 coating thickness with and without Fe micrograins.

thicknesses, more Fe micrograins that are randomly dispersed on the surface are found. The surface roughness of the samples was further investigated using a surface texture tester and the results are shown in Fig. 4. The surface roughness increases significantly on the thicker coatings. Furthermore, the MAO Al_2O_3 coatings embedded with Fe micrograins show a coarser surface for the same thickness. The changes in the surface morphology also indicate that many Fe micrograins are embedded in the Al_2O_3 coatings through diffusion and electrophoresis, and the coatings are much denser compared to those without Fe micrograins.

After Fe micrograins incorporation, the samples exhibit increased microhardness, as shown in Fig. 5. It should be noted that accurate measurement of the microhardness on the 50 µm thick coating is difficult because the coating is rather coarse. Nonetheless, our study suggests that the increased hardness is due to the decreased porosity and consequently denser surface. Fig. 6 depicts the friction coefficients μ of the base materials (uncoated Al alloy as reference) and MAO Al_2O_3 coatings of different thicknesses with and without Fe micrograins measured against a WC ball. The friction coefficients of the MAO samples decrease obviously. In addition, the friction

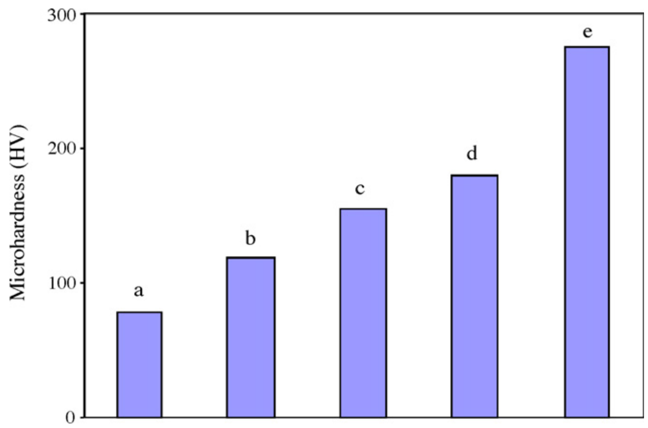

Fig. 5. Microhardness of the untreated sample and MAO Al_2O_3 coated samples at a load of 200 g: (a) untreated sample; (b) 5 µm coating without Fe; (c) 5 µm coating with Fe; (d) 20 µm coating without Fe; (f) 20 µm coating with Fe.

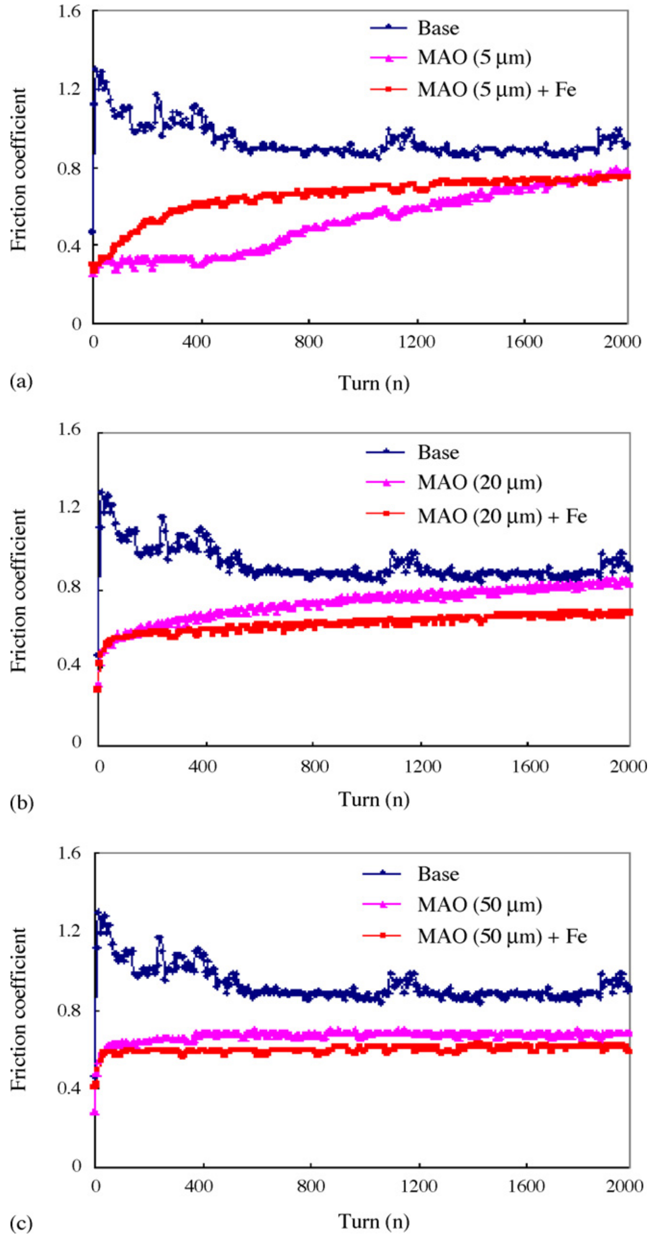

Fig. 6. Friction coefficients as a function of sliding turns measured from the Al-alloy and MAO Al_2O_3 coatings of different thicknesses with and without Fe micrograins: (a) 5 µm coatings; (b) 20 µm coatings; (c) 50 µm coatings.

coefficients of the MAO Al_2O_3 samples with Fe micrograins are more stable, and the values are slightly lower than those without Fe micrograins at thicknesses of 20 and 50 µm (0.8–0.65 and 0.7–0.6, respectively). All the coatings show insignificant weight loss after the sliding tests. In order to better understand the wear behavior of the MAO Al_2O_3 coatings with and without Fe micrograins, the wear tracks were evaluated using optical microscopy (50× for the untreated samples and 100× magnification for the coated samples). The micrograpghs are displayed in Fig. 7. The wear tracks of the MAO Al_2O_3 coatings with Fe micrograins are narrower and more compact compared to those on the MAO Al_2O_3 coatings without Fe micrograins of the same thickness.

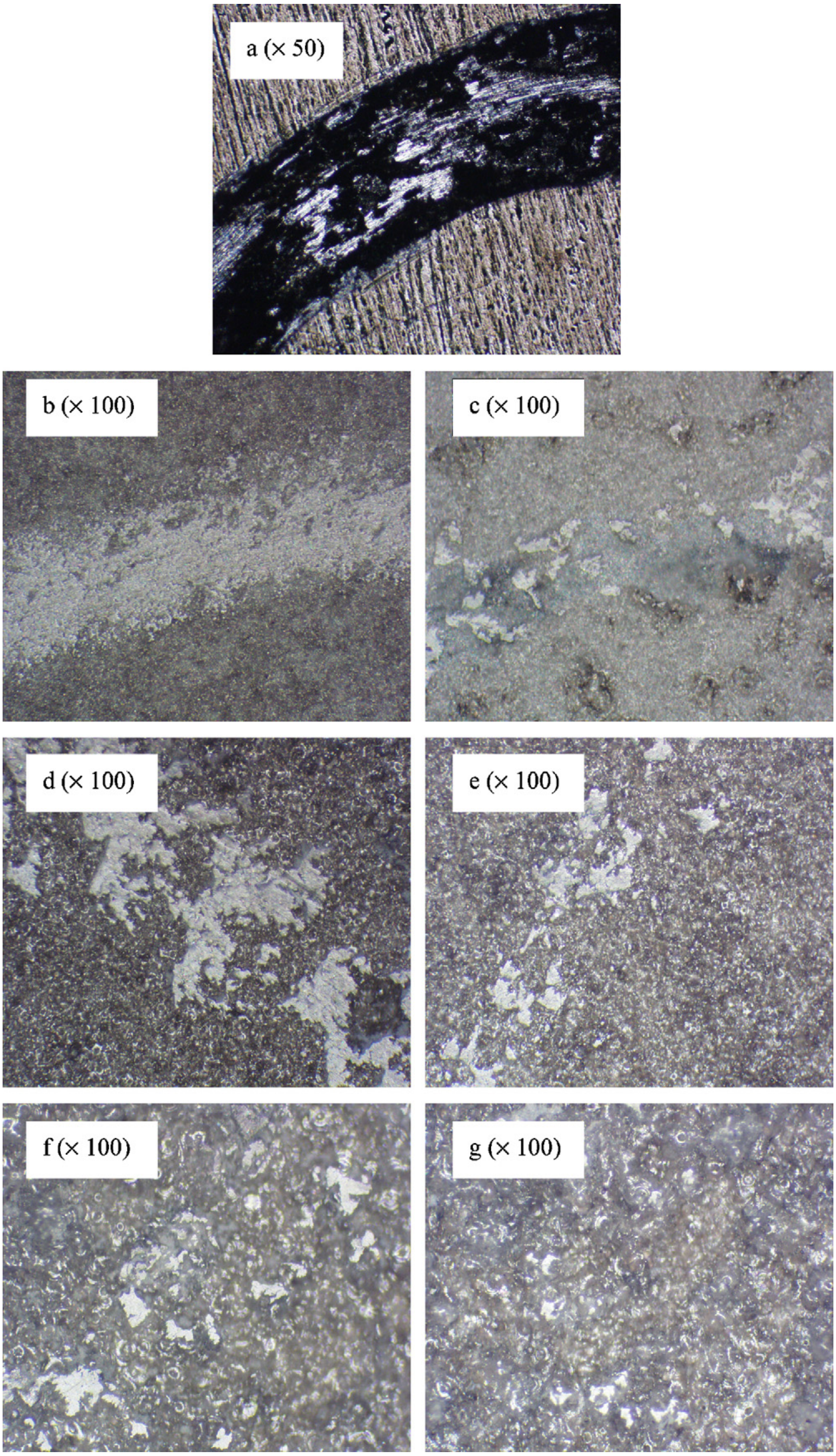

Fig. 7. Wear tracks: (a) untreated sample; (b) 5 μm coating without Fe; (c) 5 μm coating with Fe; (d) 20 μm coating without Fe; (e) 20 μm coating with Fe; (f) 50 μm coating without Fe; (g) 50 μm coating with Fe.

Previous reports have indicated different wear mechanisms for MAO Al_2O_3 coatings with different compositions during the sliding wear tests, and the friction behavior of the oxidized samples is complicated [5]. Here, we have empirically determined that the reduction in the friction coefficient arises from the improved surface hardness and reduced porosity of the MAO Al_2O_3 coatings embedding with Fe micrograins. Our results also show that the mechanical properties of MAO coatings are closely related to the surface structure. While conventional MAO coatings often possess porous surfaces, introduction of Fe micrograins into the electrolyte alters the structure by possible reactions occurring in the micro-arc discharge channels such as diffusion and electrophoresis during MAO. Consequently, a denser and less porous MAO Al_2O_3 coating structure is attained.

4. Conclusion

Al_2O_3 coatings with and without embedded Fe micrograins of different thicknesses were fabricated on Al alloy by micro-arc oxidation. Incorporation of Fe was accomplished by adding Fe micrograins into the electrolyte in MAO. Our results show that Fe micrograins are successfully embedded in the Al_2O_3 coatings and the embedded coatings are denser and have lower porosity. The mechanical properties of the Fe-embedded MAO Al_2O_3 coatings such as hardness and wear resistance are consequently improved. Our results also show that the mechanical properties of MAO coatings are closely related to the surface structure. Introduction of Fe micrograins into the electrolyte is believed to alter the structure of the films because of reactions occurring in the micro-arc discharge channels such as diffusion and electrophoresis.

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