Wear 342-343 (2015) 172–180

Contents lists available at ScienceDirect

Wear

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

# Effect of graphite concentration on the friction and wear of Ni–$Al_2O_3$/graphite composite coatings by a combination of electrophoresis and electrodeposition

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A R T I C L E  I N F O

*Article history:*
Received 8 February 2015
Received in revised form
25 August 2015
Accepted 27 August 2015
Available online 5 September 2015

*Keywords:*
Non-ferrous metals
Other surface engineering processes (electrophoresis–electrodeposition)
Metal-matrix composite (Ni–$Al_2O_3$/graphite composite coatings)
Lubricant additives
Hardness
Wear testing

A B S T R A C T

The purpose of this work was to investigate the sliding friction and wear of a novel Ni-based coating. Ni–$Al_2O_3$/graphite composite coatings were prepared on LY12 aluminum alloys using a three-step process that involved electrophoresis and electrodeposition. Uniform composite coatings with a high volume content of particles were prepared. The effects of the deposition process on the wear mechanism of these coatings were investigated. The microindentation hardness decreased, while the friction coefficient and wear loss of the coatings initially decreased and then slowly increased as the graphite content was increased. The optimum value of the graphite particle concentration in the electrophoresis bath was 1.5 g/L. The embedded graphite particles formed a transfer film on the contact area and reduced the sliding friction coefficient of the composite coatings. The new Ni–$Al_2O_3$/graphite composite coating presented excellent lubricating properties and wear resistance largely due to the effects of graphite and $Al_2O_3$ particles.



## 1. Introduction

Aluminum alloys have been widely used in engineering like architecture, automobile, military, navigation, aviation and aerospace, due to their good thermal conductivity, light weight and good mechanical strength and corrosion resistance [1,2]. Regrettably, friction coefficient and wear rate of the aluminum alloy are very high under dry sliding. Consequently, indispensable methods are needed to improve the tribological properties of the aluminum alloys for many special applications.

Composite coatings are widely applied to mechanical components due to their high mechanical, chemical and tribological properties, as well as excellent corrosion resistance [3–5]. For examples, Jiang et al. [6] prepared three CrFeAlTi composite coatings on 304 stainless steel via the combined techniques of thermal spraying and subsequent laser sintering, they found that the high temperature oxidation and corrosion resistances are better than that of the substrate. Das et al. prepared [7] electroless nickel composite coating with 2.9 wt% $Si_3N_4$ on ferrous based bearings which could be suitably used for water lubricated applications, and showed satisfactory performances in terms of weight loss, pitting levels and coefficient of friction and life time of about 9 years under the actual application environment. Sun et al. [8] fabricated (Ti, O)/Ti or (Ti, O, N)/Ti coatings by the plasma immersion ion implantation and deposition (PIIID) technique on a NiTi shape memory alloy (SMA, 50.8 at% Ni), and found that the width of wear tracks on (Ti, O, N)/Ti coated NiTi SMA samples was reduced 6.5-fold, in comparison with that on uncoated samples, demonstrating an improved wear resistance.

Furthermore, composite coatings containing fine abrasive particles like silicon carbide (SiC), alumina ($Al_2O_3$), silicon nitride ($Si_3N_4$), diamond etc. can be remarkable to increase the hardness and reduce the friction coefficient as well as the wear rate of the mechanical components [9–12]. However, the volumetric content of particles in the composite coatings prepared with the traditional electroplating and electroless plating is not high enough to satisfy the need of actual application in some special fields.

In recent years, it has aroused a growing interest in the development of using the solid lubricant particles to form a self-lubricating film [13–15]. And graphite materials have excellent lubricating properties due to the intrinsic layered structure and corrosion resistance, they are commonly used to prepare self-lubricating composite coatings [16]. Wu et al. deposited graphite particles into the nickel matrix by electroplating, and found that

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http://dx.doi.org/10.1016/j.wear.2015.08.020
0043-1648/

the wear resistance and anti-friction of the composite coatings with graphite particles were better than that of composite coatings without graphite particles [17].

Electrophoretic deposition (EPD), as an effective and economical method of materials processing technique for widely applications, is extremely attractive. EPD has been successfully applied to several traditional and advanced materials [18]. EPD process often involves two main steps: charged particles suspended in solution move toward the oppositely charged electrode under the electric field, with particles depositing on the electrode surface in the first step, and then form an uniform film in the second step. For the purpose of producing metal/ceramic composite coatings, EPD has been increasingly employed in combination with electroplating or electroless plating, which could largely improve the wear resistance of materials [19,20]. Shrestha successfully produced composite coatings of nickel and ceramic particles in two steps (EPD and electroplating), and the composite coatings showed higher volumetric content of ceramic particles and better anti-wear performance than those of the composite coatings fabricated with the single step (electroplating) [21].

This work was devoted to fabricate the anti-wear and self-lubricating composite coatings on aluminum alloys with alumina and graphite particles in three steps by a combination of electrophoresis and electrodeposition, due to the good self-lubricating property of graphite that is conductive to reduce the friction coefficient of the composite coatings [16,17], and the high melting point, high hardness, high rigidity and the excellent chemical stability of $Al_2O_3$, which help to improve the hardness and thus the wear resistance of the composite coatings [3,10,13,20–22]. This work mainly concentrated on increasing volumetric content of particles into the nickel matrix and the study of the wear behavior of the composite coatings. Firstly, a thin nickel was electrodeposited on the substrate (Nickel pre-plating) because the direct electrophoresis deposition on the aluminum alloys is not easy, and then alumina and graphite particles were deposited onto the thin nickel by the EPD method, followed by another nickel electrodepositing. The hardness and wear resistance of the coating would be enhanced with the distributing of a high volumetric content of alumina particles. However, graphite particles, which serve as solid lubricant, may have an adverse effect on the hardness of the coatings. Thus, the composite coatings were prepared with different concentrations of graphite particles in the EPD bath, and the friction and wear behavior of these coatings under dry sliding conditions was studied. Additionally, the optimum graphite particles concentration was obtained.

## 2. Experimental details

### 2.1. Nickel pre-plating

An aluminum alloy (LY12) substrate (15 mm × 12 mm × 8 mm) was polished with $2000^{\#}$ abrasive paper and cleaned by ultrasonic vibration with acetone for 15 min, and then rinsed by the distilled water. After drying the substrate in air, it was placed vertically and paralleled to a nickel anode plate (99.9% in purity) at a distance of 2–3 cm. Chemical composition of the electrodeposition plating bath and the electroplating process parameters are listed in Table 1.

**Table 1**
Electroplating bath composition and operating conditions.

| Composition and operating conditions | Values |
|---|---|
| $NiSO_4 \cdot 6H_2O$ (nickel sulfate hexahydrate) | 300 g/L |
| $NiCl_2 \cdot 6H_2O$ (nickel chloride hexahydrate) | 20 g/L |
| $H_3BO_3$ (boric acid) | 50 g/L |
| Temperature | 60 °C |
| Duty ratio | 60% |
| Voltage (cell-potential) | 1.75 V |
| Plating time | 3 min |
| pH | $4.0 \pm 0.2$ |

### 2.2. Electrophoretic deposition

A 50 mm × 20 mm × 5 mm graphite plate was used as the anode during the electrophoretic deposition. Two kinds of particles were applied to this process: $Al_2O_3$ (α-type, average size 150 nm, ST-NANO, China) and graphite (average size 1 μm, Qingdao Senyan Graphite Co., Ltd., China) particles. The electrophoretic deposition was carried out on the pre-deposited nickel matrix by using an ethanol bath containing 1.5 g/L of $MgCl_2 \cdot 6H_2O$ and a suspension of 12 g/L of $Al_2O_3$ particles, where the graphite particles concentrations were 0, 0.5 g/L, 1.0 g/L, 1.5 g/L, 2.0 g/L and 2.5 g/L in the EPD bath. Meanwhile, the bath was stirred by a magnetic stirrer at a speed of 150 rpm for 30 min and then ultrasonicated for 1 h by using an ultrasonic cleaner before the electrophoretic deposition. This process was performed under an electric voltage of 40 V for 250 s at room temperature. 10–20 mL $C_6H_{15}NO_3$ which act as dispersing agent was added into the electrophoretic deposition bath, where the ethanol solution acted as dispersion medium, so as to prevent the agglomeration of the graphite and alumina particles, with the combined help of the mechanical stirring and ultrasonic dispersion. During the deposition, the EPD bath was stirred by a magnetic stirrer at a constant speed of approximately 150 rpm.

### 2.3. Electrodeposition of nickel

The electrodeposition of nickel was carried out by using pulse power. Two 70 mm × 20 mm × 5 mm nickel plates were used as anodes, and the substrate which was covered with the electrophoretic coating was used as cathode. The pulse electrodeposition was carried out in a Watt bath containing 300 g/L $NiSO_4 \cdot 6H_2O$, 70 g/L $NiCl_2 \cdot 6H_2O$, and 10 g/L $H_3BO_3$, at a potential of 5 V and a pulse duty ratio of 60% for 2 h, the bath was maintained at 60 °C without stirring, and the bath pH was adjusted at 4.0–5.0.

At last, the samples with composite depositions were cleaned by ultrasonic vibration with acetone for 3 min and then rinsed by the distilled water, to remove the excess particles that were loosely adsorbed to the surface. The whole process could be concluded as the general scheme [21] which shows the three-step method of composite coating, as plotted in Fig. 1.

### 2.4. Analysis of composite coatings

After the composite depositing, the samples were washed in running water and then sonicated at an ultrasonic frequency of 20 kHz in acetone for 5 min. The surface and cross-section micrographs of the composite coatings were examined by using scanning electron microscope (SEM, JSM-6390A, Japan). The composition and the volumetric concentration of particles in composite coatings was determined by using X-ray diffraction (XRD, XRD-6000, Japan) and energy dispersive spectroscopy (EDS) coupled to the SEM. As the O element in the air may affect the real content of the O content in the composite coating, the Al element is chosen as the basic element of the volume fraction calculation for $Al_2O_3$ particles. Comparing with the above-mentioned EDS measurements, the volume fraction of the $Al_2O_3$ and graphite particles in the composite coating can be calculated according to