Electrodeposition of Ni-Nitride composite coatings: A review of recent study

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Abstract. This work point to a wide potentially on performance improvement of transition metal-based coating system especially Ni-TiN-AlN and Ni-TiN-AlN/Si3N4 composite coatings by using electrodeposition process. Electrodeposition is widely used in metal matrix composite for many industrial applications due to a relatively cheap solution to enhance the composite performance. The aim of this work is to study the influence of various current density and nitride particles concentration on corrosion behaviour of composite coatings. The corrosion test was performed by immersing the deposited composite samples into acid solution for 2 hours. The results showed that the corrosion rate of Ni-TiN-AlN and Ni-TiN-AlN/Si3N4 composite coatings tend to increase as the current density was increased. While, the corrosion rate of Ni-TiN-AlN composite coatings at various nitride (TiN, AlN) particles concentration tend to decrease as the nitride (TiN, AlN) particles concentration was increased. It was shown that corrosion resistance of Ni-TiN-AlN/Si3N4 composite coatings at various Si3N4 was excellent due to the effectively incorporation of the Si3N4 particles as the corrosion barrier.

1. Introduction

Thin film (coating) deposition is a new trend for many industrial applications such as electronic, energy and mechanical properties. The impact of coating development is to expand the material or tool life through the improvement of coating properties [1]. Nickel (Ni) thin film was widely used as protective coating due to its excellent physical and mechanical properties. Although it has limited on wear and corrosion resistance especially at high temperature, however nickel composite system (combination of nickel and particles) can improved its performance [2-4].

Meanwhile, the transition metal nitride particles of TiN, AlN and Si3N4 compound commonly used as reinforced particle to improve the wear and corrosion resistance of composite material [5-7]. Furthermore, the combination of TiN and AlN can be formed as a multilayer TiN/AlN composite that significantly improve its mechanical and corrosion properties [8,9]. Moreover, the combination of TiN and Si3N4 can be formed as a super hard TiN/Si3N4 composite as a nanostructured material [10]. The composite retained its high hardness at high temperature operation.

The electrodeposition is an established deposition technique in developing metal matrix composite coating [11]. The technique has a superiority in producing composite as well as nanostructured materials with a wide variety of materials (including Ni) and applications [12].
The properties of electrodeposited composite coating is strongly influenced by working parameters such as current density, particles concentration and temperature etc. [3,13-15]. The current density is one of the important parameter since it control the ionization of metal element during electrodeposition process. While the compound particles that are inert particles such as nitrides can be entrapped in metal matrix by electrophoresis that is controlled by current density. In electrodeposition process, the increase of particles concentration give the effect of agglomeration and sedimentation.

In this paper, a brief analysis on corrosion behaviour of the electrodeposited nickel-nitride (Ni-TiN-AlN and Ni-TiN-AlN/Si3N4) composite coatings from previous study is presented. The effect of electrodeposition parameters in term of current density and nitride particles concentration on the corrosion rate in term of mass loss rate of the composite coatings are discussed.

2. Experimental investigation

2.1. Experiment of Ni-TiN-AlN composite coating
The experimental investigation were conducted on electrodeposition of Ni-TiN-AlN composite coating [16,17]. The electrodeposition process was carried out by using potensiostat system (Edaq Z100) with three electrode mounting containing a platinum (Pt) wire as the counter electrode, AgCl wire as reference electrode and tungsten carbide bar (dia. 1 mm) as working electrode (substrate). The electrodeposition was performed from distilled aqueous solution containing 0.38 M nickel sulfate hydroxide (NiSO4·6H2O), 0.17 M nickel chloride hydroxide (NiCl2·6H2O) and 0.49 M boric acid (H3BO3) mixed with titanium nitride (TiN) and aluminum nitride (AlN) powder by using double-distilled aquades and then magnetically stirred for about 24 hours. In the first study, the electrodeposition process of Ni-TiN-AlN composite coating was carried out by varying current density of 6, 9 and 12 mA/cm² for 15 minutes with fixed TiN and AlN concentration of 2 g/L. In the second study, Ni-TiN-AlN composite coatings were deposited at fixed current density of 6 mA/cm² and various TiN and AlN particles concentration of 4, 6 and 8 g/L.

2.2. Experiment of Ni-TiN-AlN/Si3N4 composite coating
Further study was done on electrodeposition of Ni-TiN-AlN composite coating combined with Si3N4 particles to form Ni-TiN-AlN/Si3N4 composite coating system [18,19]. The bath composition was 0.38 M NiSO4·6H2O, 0.17 M NiCl2·6H2O, 0.49 mol/L H3BO3, 1.2 g/L SDS, 4 g/L TiN and 4 g/L AlN. First experiment was performed at fixed current density of 2 mA/cm² for 15 minutes and various Si3N4 particle concentration of 0.2, 0.4 and 0.6 g/L. Pt and AgCl wire were used as counter and reference electrode, respectively. Tungsten carbide bars (0,42x0,42x3,82) cm were employed as working electrode as well as substrate. Second study was carried out by electrodepositing Ni-TiN-AlN/Si3N4 composite coating at various current density of 1.5, 2 and 2.5 mA/cm² [20,21]. The bath composition was 0.38 M NiSO4·6H2O, 0.17 M NiCl2·6H2O, 0.49 mol/L H3BO3, 1.2 g/L SDS, 2 g/L TiN, 2 g/L AlN and 10 g/L Si3N4.

2.3. Corrosion test
Simple corrosion test were conducted by immersing the samples of Ni-TiN-AlN composite coatings into 3.5% HCl solution for 2 hours. While the samples of Ni-TiN-AlN/Si3N4 composite coating were immersed into 1 M H2SO4 solution for 2 hours. All the samples mass before and after immersion were measured and the mass loss rate of the all composite samples after immersing in strong acid solution were calculated.

3. Results and discussion

3.1. Ni-TiN-AlN composite coating
The corrosion test result, in term of mass loss, of Ni-TiN-AlN composite coating at different current density and nitride particles are shown in Figure 1 and Figure 2, respectively.
For the first study, it shows that the mass loss rate of composite coatings tend to increase as the current density is increased up to 9 mA/cm² and it decreases as the current density is further increased. Meanwhile, for the second study, the mass loss is decreased as the nitride (TiN and AlN) particles concentration is increased up to 8 g/L. It was reported that the reinforced particles gave a better effect in improving the corrosion resistance due to the reduce of hole or pore size of nickel composite morphology [18]. It was shown that the surface morphology of Ni-TiN-AlN composite coatings appeared rough surface with large separation between grains as the current was increased [16]. Thus it provided the sample to be easy attacked by acid solution leading high mass loss rate. However, the surface morphology of coating became finer as further increasing in current and lead to the decrease of mass loss rate.

From this result, it obviously showed that the particles concentration as well as current density was a critical parameter in electrodeposition process. The coating surface is refined as the nitride particle concentration is increased [14] and it is also confirmed in this study leading the decrease of mass loss rate. The surface morphology also was determined by the existence of metal elements of coating. It was observed that the metal element of Ni was obtained but not Ti and Al elements for the coating deposited
at nitride particles concentration of 4 g/L. As the particles concentration was increased, all metal elements such as Ni, Ti and Al were revealed resulting finer morphology [16]. Meanwhile, the current density also influenced the coating composition. The lower current density lead the lower number of metal particles reach the surface cathode due to less metal ionic [17] and it cause the rougher surface morphology.

3.2. Ni-TiN-AlN/Si$_3$N$_4$ composite coating

The study results of the corrosion rate of Ni-TiN-AlN/Si$_3$N$_4$ composite coating at various Si$_3$N$_4$ particles concentration and various current density are shown in Table 1 and Figure 3, respectively. The result shows that Ni-TiN-AlN/Si$_3$N$_4$ composite coating deposited at various Si$_3$N$_4$ particles concentration has excellent corrosion resistance with no mass loss as presented in Table 1. The addition of Si$_3$N$_4$ particles in the composite significantly improved the corrosion resistance by filling the surface defect or incorporating the Si$_3$N$_4$ particles into the composite [20]. It was shown that the incorporation of Si$_3$N$_4$ particles into composite coating resulted grain refinement [22]. The grain refinement was due to the formation nucleation site and the site number increased as the Si$_3$N$_4$ particles concentration was increased [20]. Meanwhile, the corrosion test results of Ni-TiN-AlN/Si$_3$N$_4$ composite coating at various current density showed that the increase of current density led the increase of mass loss rate and it decreased as further increasing in current density. The result is similar with prior finding.

**Table 1.** Mass loss rate of Ni-TiN-AlN/Si$_3$N$_4$ composite coating deposited at various Si$_3$N$_4$ particle concentration.

| Si$_3$N$_4$ concentration (g/L) | m$_1$ (gram) | m$_2$ (gram) | Δm (gram) |
|-------------------------------|-------------|-------------|----------|
| 0,2                           | 8,744       | 8,744       | 0,000    |
| 0,4                           | 9,230       | 9,230       | 0,000    |
| 0,6                           | 9,156       | 9,156       | 0,000    |

**Figure 3.** Mass loss rate of Ni-TiN-AlN/Si$_3$N$_4$ composite coating deposited at various current density.

4. Conclusion

A recent study on the properties of Ni-Nitride composite coatings, especially the influence of current density and nitride particles concentration on its corrosion behaviour in term of mass loss rate has been reported. Generally, the mass loss rate of Ni-TiN-AlN and Ni-TiN-AlN/Si$_3$N$_4$ composite coatings increased as the current density was increased. Meanwhile, the mass loss of Ni-TiN-AlN composite coatings decreased as the nitride (TiN, AlN) particles concentration was further increased. It was found
that the corrosion resistance of Ni-TiN-AlN/Si3N4 composite coating at various Si3N4 particles concentration was excellent. It was concluded that the nitride particles especially Si3N4 particles played the role of corrosion barrier in nickel-/nitride composite coating.

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