MORPHOLOGY OF Ni-TiN/Si_3N_4 COMPOSITE COATINGS AT HIGH-TEMPERATURE OXIDATION

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ABSTRACT

Electrodeposited nickel coating has excellent physical and mechanical properties, however it does not withstand to high temperature oxidation for long time. The addition of nitride phase within the nickel based coating improve the oxidation resistance through dense surface morphology improvement. A preliminary study was performed to investigate the high temperature oxidation behavior on the coating morphology after exposed at high temperature. The Ni-TiN/Si_3N_4 composite coatings deposited at electrodeposition temperature of 35 °C, 40 °C and 50 °C. The electrolyte consists of 0.17 M $NiCl_2.6H_2O$, 0.38 M $Ni_2SO_4.6H_2O$, 6 g/L TiN, 0,6 g/L Si_3N_4, 40 g/L H_3BO_3, 0,6 g/L *Sodium Dodecyl Sulfate* (SDS). High temperature oxidation process on the samples was performed by heating at temperature of 700 °C for 2 hours within air conditions. The morphology and composition samples were characterized by using SEM/EDS. In general, the result showed that the morphology of Ni-TiN/Si_3N_4 composite coatings became rough due to the oxidation process. Elements coating such as Ni, Ti, Si and N were disappeared after the oxidation process due to the outward diffusion process.

Keywords: Composite; Electrodeposition; Morphology; Oxidation

Introduction

Oxidation resistance of composite coating especially at high temperature operation is an important characteristic for its wider applications. Nickel (Ni) material has been used as coating due to its resistance to wear and corrosion.[1-2] However, at high temperature operation above 500 ^{0}C, the nickel layer would be oxidized due to the oxygen diffusion into the layer.[3] To prevent the oxidation process of nickel, some reinforcement particles such as nitrides were added to form Ni-based composite coating. Titanium (Ti) and Silicone (Si) had significant contribution in improving anti-corrosion and wear.[4,5] The combination of Ti and Si with nitrogen (N) in forming a nitride compound of TiN and Si_3N_4, respectively, has improved their properties and led their role as reinforced particles within the Ni-based composite coating.[3,6]

High temperature oxidation resistance at 650 – 950 ^{0}C has been concerned as an interest challenge for composite coating technology.[7,8] Pure Ni with porous surface morphology may be oxidized at temperature of 700 ^{0}C.[9,10] At low temperature, the oxidation is resisted by forming NiO layer,[11] however at high temperature the inward diffusion of oxygen (O) into the coating become massive and oxidized the substrate. Some nitride particles were added into Ni coating to improve its oxidation resistance through modification of its surface morphology. The modification includes elimination of grains interspaces, hinder the crystal growth and increasing coating density.[12-14]

The smooth and homogeneous surface morphology of composite coating was influenced by crystal growth that was driven by process temperature.[15,16] In

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electrodeposition process of nickel composite coating, the temperature increase in 30-60 °C improved the mobility of electrolytes molecule or particles since it degraded the electrolyte viscosity. It also influenced the adatom energy led to the increase of diffusion and particle mobility onto the surface. The previous investigation result showed that the optimum electrodeposition temperature was 40 °C due to the lowest wear rate of the coating.[17]

In the present study, analysis on Ni-composite coating is subjected to investigate the effect of high temperature oxidation on the surface morphology and composition of Ni-TiN/Si_3N_4 composite coating. The discussion on EDS composition analysis is presented.

Methods

Ni-TiN/Si_3N_4 composite coatings were deposited on tungsten carbide plate (4x4x38 mm) by electrodeposition process for 15 minutes at fixed current density of 2.5 mA/cm^2.[18] The electrodeposition temperature variation was applied at 35 °C (sample 1), 40°C (sample 2) and 45 °C (sample 3). 5 ml of electrolyte solution was composed of 0.17 M $NiCl_2.6H_2O$, 0.38 M $Ni_2SO_4.6H_2O$, 6 g/L TiN, 0,6 g/L Si_3N_4, 40 g/L H_3BO_3, 0,6 g/L *Sodium Dodecyl Sulfate* (SDS). The electrodeposition cell consisted of tungsten carbide plate as working electrode (substrate), Platinum (Pt) wire as counter electrode and AgCl wire as reference electrode, respectively. During the process, the electrolyte was magnetically stirred to disperse the nitride particles. The oxidation process for all samples was carried out by using furnace at constant temperature of 700 °C for 2 hours in atmosphere condition. The morphology of samples was characterized by using Scanning Electron Microscopy (SEM) while the composition is analyzed by Energy Dispersive Spectroscopy (EDS).

Result and Discussion

SEM surface morphology of the Ni-TiN/Si_3N_4 electrodeposition composite coating after high-temperature oxidation process at 700 ℃ is presented in Figure 1. The sample 1 shows an even morphology with large protuberances while the others show sharp protuberances. From EDS result, the large protuberances of sample 1 was resulted from even large grain size of Ni while the others, there were no coating elements were detected as seen in Table 1. Ti, Si and N are depleted and disappeared due to outward diffusion at high temperature oxidation.[19,20]

From the result of sample 1, it shows that Ni is successfully utilized as protective element coating as the material exposure at high temperature. The oxidation surface of coating can be explained by dissociative mechanism model which at high temperature, the growth stress is generated within the oxide and it can cause the void formation at interface and eventually forming the porous zone between coating layer and substrate.[19]

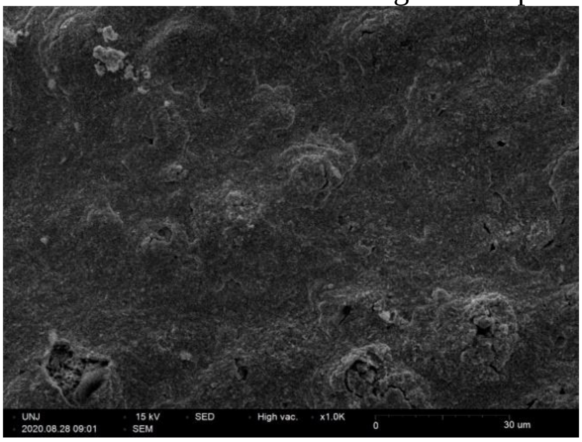
The high Ni metal activity at interface cause the cation migrate outward and eventually decrease the metal activity itself followed by rising oxygen activity into pore and form oxide of NiO at interface.

Meanwhile, according to experimental result,[20] Si element play role in nanostructure formation with amorphous silicon nitride formation at grain boundary, grain refinement and densification, retarded the formation of pores due to nitrogen release, formation of stable oxide SiO avoiding oxygen diffusion into the substrate. The rough morphology of oxidized samples as long exposure time is due to thermodynamic differences of oxidation product causes the irregular oxide/alloy interface.[21]

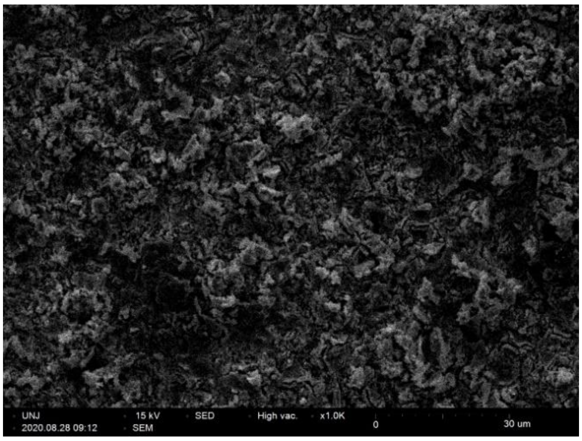
It was also reported that the rough surface of Ti alloy was ascribed to lattice mismatch between oxide layer and substrate.[22] Furthermore, from TiN coating experimental results, it showed that at high temperature of 450 ^{0}C the nitrogen is replaced by oxygen and form oxide layer on the top of surface[23] it always presence on the outer zone of the coating.[24]

It is observed that during the oxidation process, N diffuse outward while O diffuse inward. At the surface, TiN is oxidized into TiO while Si_3N_4 is oxidized into SiO_2.[25]

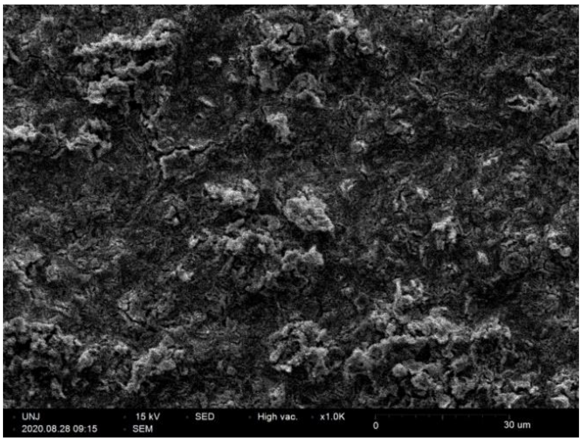
Meanwhile, Ni tends to accumulate at the surface. However, at higher temperature oxidation as in this study, the oxidation rate is much increased cause depletion of Ti, Si and N elements until zero including Ni except for sample 1. Compare with two other samples, befor oxidation process, Ni content of sample 1 is the highest.



(a)



(b)



(c)

Figure 1. SEM morphology of Ni-TiN/Si_3N_4 composite coating electrodeposited at temperature of (a) 35 °C, (b) 40 °C and 45 °C. All coating samples were exposed at temperature of 700 °C for 2 hours within air condition

Table 1. Ni-TiN/Si_3N_4 Composition before (a) and after (b) oxidation at 700 °C

(a) Before oxidation process

Sample	***At% Element***					
	O	***W***	***Ni***	***Ti***	***Si***	***N***
1	65,11	0,45	14,82	0,73	1,05	17,84
2	62,39	-	13,9	1,09	1,87	20,74
3	67,12	11,38	7,42	0,14	2,24	11,71

(b) After oxidation process

Sample	***At% Element***					
	O	***W***	***Ni***	***Ti***	***Si***	***N***
1	44,8	39,83	13,36	-	-	-
2	47,44	51,29	-	-	-	-
3	48,42	46,44	-	-	-	-

Conclusion

The behavior of Ni-TiN/Si_3N_4 composite coating at high temperature oxidation of temperature of 700 ℃ has been studied. It showed that the surface morphology became rough after the oxidation process especially for sample 2 dan sample 3. Meanwhile, the sample 1 showed a smoother surface morphology than others. From the composition analysis on the coating element, it showed that Ni, Ti, Si, N were disappeared due to the outward diffusion. However, only the sample 1 showed the rest of Ni element after the oxidation process due to its highest Ni content of initial composition before the process compare to two other samples.

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