

# Tribology performance of CrAlSiN coatings deposited by pulsed current input cathodic arc evaporation

Wei-Che Huang<sup>1</sup>, Cheng-Hsun Hsu<sup>1</sup>, Wei-Yu Ho<sup>2</sup>, Chien-Liang Lin<sup>2,\*</sup>

<sup>1</sup> Department of Materials Engineering, Tatung University, Taipei, 104, Taiwan

<sup>2</sup> Department of Materials and Energy Engineering, MingDao University, ChungHua, 523, Taiwan

\*Corresponding author: cllin@mdu.edu.tw

**Abstract.** This study has explored the effect of cathodic arc evaporation technology using pulsed current input on the wear resistance of the CrAlSiN coatings. The input of arc source current was set by modulating arc pulsed current in the two ranges of 90/120 A as well as 90/150 A, respectively; and also set a constant current of 90 A for comparison. These different current inputting conditions were controlled to obtain three kinds of CrAlSiN coatings in cathodic arc deposition system. The results show that all the coatings have the specific B1-Rock salt structure indexed by their XRD patterns with the (111) and (200) diffraction peaks. The coatings deposited with the pulsed current as compared to that with constant current presents the higher hardness up 4145 HV. After the TGA/DSC analysis in atmosphere, all the coatings are stable up to 1000 °C. The result of tribology test also shows that the CrAlSiN coating deposited with the pulsed current of 90/150 A has the best wear resistance.

## 1. Introduction

In order to search new coating materials and coating synthesis processes, many studies are under increasing demands to improve the properties of the proper coatings for the cutting tool with better efficiency. In the past decades, with unique nano-composite structure, CrAlSiN coatings have achieved an acceptance in various fields of machining applications due to the advantage of excellent mechanical properties [1-5]. For instance, CrAlSiN coating is widely applied to the various cutting tools featured with excellent property of high hardness up to 42 GPa. Furthermore, CrAlSiN has been proven with attractive oxidation resistance because some protective phases such as Al<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> generate on its surface to retard the diffusion of oxygen into the coating. In a previous study [6], the CrAlSiN film was coated on tool steel and the result showed a slightly lower friction coefficient as compared to uncoated one. In view of this result, we are interested in the exploration about the more effective performance of wear resistance of CrAlSiN.

The pulsed cathodic arc deposition technology is one of the popular PVD techniques to synthesize hard coatings on cutting tools [7-12]. Based on this technique, a novel concept with the advantages of pulsed current input of the sources has been developed with the purpose of improved the coating systems in comparison with conventional un-pulsed arc depositing coatings. By means of the pulsed current supply for the cathodic arc sources, evaporation flux of metallic ions can be intensified without further modifying the arc sources. Therefore, the present work mainly aims to present the performance of CrAlSiN coatings obtained by using the above-mentioned technology, and then explore the effect of inputting pulsed current to the evaporators on improving coating wear properties.



## 2. Experimental

In the study, cathodic arc evaporation technology was used to deposit nano-composite CrAlSiN coatings on the substrates. The substrates were made of both tungsten carbide and AISI 304 stainless steel with the disc dimension of  $\Phi 20 \times 6$  mm in size. The coating system consisted of two sets of standard cathodic arc evaporators, adding a circular substrate holder to connect with bias power supply. Two targets, chromium metal (purity: 99.9%) and  $\text{Cr}_{30}\text{Al}_{60}\text{Si}_{10}$  alloy, were used as source materials, respectively. The  $\text{N}_2$  and argon gas introduced into the chamber were set by the ratio of 1:1. The arc source input was set by modulating the current with the two different pulsed ranges of 90/120 A and 90/150 A, respectively. The duty cycle of the pulsed current was set at 2 milliseconds. Another condition was also used a constant current of 90 A to coat with specimen for comparison. Finally, three CrAlSiN coatings were obtained by varying the source current value, and remarked as S1 (constant 90 A), S2 (pulsed 90/120 A), and S3 (pulsed 90/150 A), respectively.

Crystallographic structure of the coatings was analyzed by using X-ray diffractometer (model: PHILIPS PANalytica X'Per PRO MRD) with a grazing incidence angle of  $2^\circ$  and Cu K $\alpha$  radiation ( $\lambda=1.54060$  Å). The surface morphology observation and coating thickness measurement was carried out by using scanning electron microscopy (FE-SEM, model: JOEL JSM-5600). Hardness of the coatings was measured using a Vicker's indentation equipment with an applied load of 25 g. Thermal analysis was performed in air using thermal gravimetric analyzer (TGA) and differential scanning calorimetry (DSC) (TGA/DSC, mode: Mettler Toledo TGA/SDAT851). Approximately 8 mg amount of sample was placed at a alumina holder and heated at the rate of 10 K/min from the ambient temperature to 1000 °C in atmosphere. Prior to testing, the equipment was calibrated for obtaining the precise percent change in the weight of sample. Wear test was conducted through the pin-on-disc method to compare the three coatings. The testing conditions were as follows: (1) no lubricant, (2) circular track with a 12 mm diameter against a 6.0 mm diameter WC ball, (3) wear speed of 0.2 m/s, (4) normal load of 5 N, (5) room temperature, and (6) 65% relative humidity. The relationship between the friction coefficient and total travel distance of 500 m was continuously recorded during the tests. Furthermore, the morphology of worn tracks on the specimens after the wear tests were observed using FE-SEM.

## 3. Results and Discussion

### 3.1 Structure, morphology, and hardness of CrAlSiN coatings

Table 1 lists the basic properties of the CrAlSiN coatings including the film thickness and roughness. The thickness of the coatings ranged from 2.8  $\mu\text{m}$  of S1 to 3.8  $\mu\text{m}$  of S2 and S3 which was dependent upon the arc source current. It is known that the higher arc source current input leads to the thicker film. That is, the result indicates that the mutative pulsed arc technique could evidently raise the deposition rate at almost the same substrate temperature [7]. X-ray diffraction analysis was carried out to compare the microstructures of the three CrAlSiN coatings, as shown in Fig. 1. As compared to S1 specimen, the diffraction patterns of the S2 and S3 ones show a similar diffraction peaks. The three coatings reveal a well-defined polycrystalline B1 (NaCl-type) FCC microstructure. The diffraction peaks of (111) and (200) preferred orientation centered at  $37.1^\circ$  and  $43.7^\circ$  are clearly observed. The similar result indicates that the film is the CrAlN structure consisting of CrN and AlN phases [6]. It is also found there are higher diffraction peaks in the S1 specimen deposited with constant current of 90A. As for the S2 and S3 CrAlSiN coatings deposited with pulsed current, the weaker peaks seems to related to the appearance of weak peak of  $\text{Si}_3\text{N}_4$  centered at  $34^\circ$  existing. According to the JCPDs and literatures [1-5], the main phase of CrAlSiN coatings was CrAlN embedded in the amorphous  $\text{Si}_3\text{N}_4$  matrix. The result reveals that the amorphous  $\text{Si}_3\text{N}_4$  matrix may transfer to ordered matrix by using pulsed current input to the arc sources.

Table 1. Basic properties of three types of CrAlSiN coatings.

Coating type	Modulating Current input (A)	Thickness ( $\mu\text{m}$ )	Roughness (Ra, $\mu\text{m}$ )	Hardness (HV <sub>0.025</sub> )
S1	constant 90	2.8	0.211	3240
S2	pulsed 90/120	3.8	0.159	3337
S3	pulsed 90/150	3.8	0.182	4145

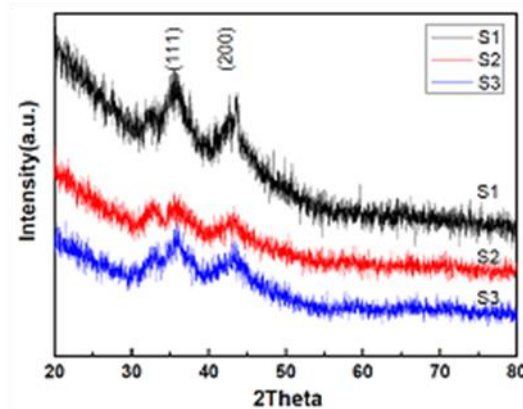


Figure 1. XRD analysis of the three CrAlSiN coatings deposited with different arc current values.

Surface morphology of the three CrAlSiN coatings examined by SEM is shown in Fig. 2. The results reveals that droplets formed during deposition from the CrAlSi cathodic target were randomly distributed on the coating surface in small sizes, varying from a fraction of a micrometer to 1  $\mu\text{m}$ . The presence of these droplets is due to the disadvantage of the cathodic arc deposition method [13,14]. It is significant that current input into the cathode could affect temperature of cathode spot and volume of melted materials. Increasing the arc current may lead to a greater number of droplets or a larger droplet size. However, from the comparison of data listed in Table 1, the surface roughness related to the droplets was decreased by using the pulsed current input, resulting from the less numbers of droplets deposited on the surface.

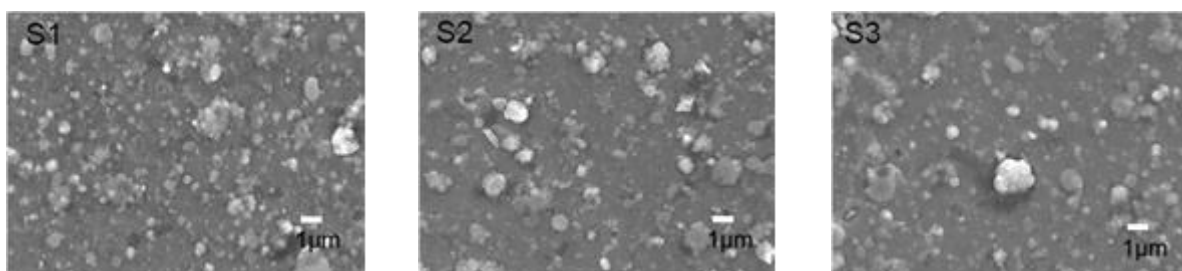


Figure 2. Surface morphologies of the CrAlSiN coatings. (a) S1, (b) S2, and (c) S3.

In addition, hardness of the three CrAlSiN coatings on tungsten carbide substrate is listed at Table 1. The hardness of the CrAlSiN coatings with different source power input shows an increase trend with increasing the arc source current input. The use of the modified pulsed arc process may result in a higher degree of ionization of the metal-plasma, and higher energy in the plasma-particles as well. When the pulsed current increases (90/120 A  $\rightarrow$  90/150 A), the hardness of the coating also increase (3337  $\rightarrow$  4145 HV), the correlation between pulsed current input and coating hardness could be clearly proved here. Particularly, the CrAlSiN coating of S3 has the highest hardness value of about 4145 HV.

### 3.2. Thermal and wear behavior of CrAlSiN coatings

Thermal behavior analysis was performed by TGA/DSC test at the temperature ranging from room temperature to 1000 °C for 40 min in air. The changes in both weight and heated behavior of specimens during the oxidation of the coatings are shown in the TGA/DSC curves of Fig. 3. The DSC curves showed an initial trend of exothermic reaction with temperature increasing to 300 °C and then turned to endothermic reaction until the end of test. Before 300 °C, CrAlSiN coatings exhibited exothermic behavior because of the release of compressive stress and the annihilation of some point defects. After 300 °C, the TGA/DSC curves showed a gradual increase in sample's weight and heat until the end of test. We found there was no extreme weight change throughout the oxidation process for CrAlSiN coatings. It implies where constant current or pulsed current was adapted to dense structure, all the coatings exhibited a gradual oxidation process occurred on their surface. After the end of heating, the coated samples changed weight ratio was only about 0.3 - 0.4 which means they almost had a similar oxidation resistance. It is expected that CrAlSiN coatings formed protective oxide layer on the surface to further retard the oxidation process. Figure 4 shows the surface morphologies of the CrAlSiN coatings after the test. The recrystallization of nano-grains was observed due to the presence of coexisting  $\text{Al}_2\text{O}_3$ ,  $\text{Cr}_2\text{O}_3$ , and  $\text{SiO}_2$  phases near the surface retarding the diffusion of oxygen into the CrAlSiN coating.

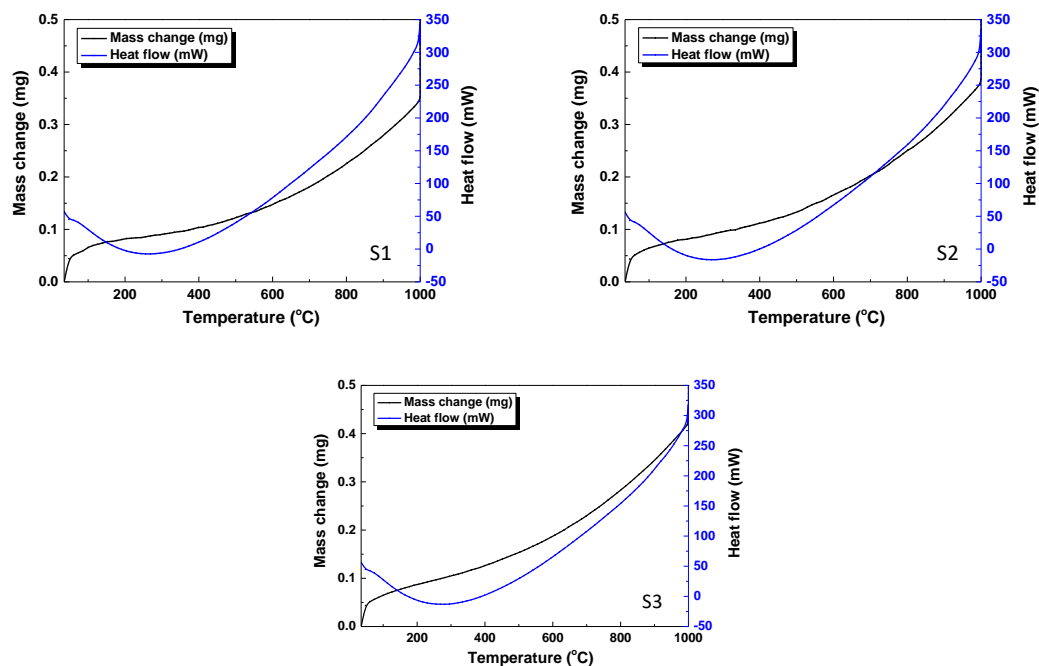


Figure 3. TGA/DSC curves of the three CrAlSiN coatings: (a) S1, (b) S2, and (c) S3.

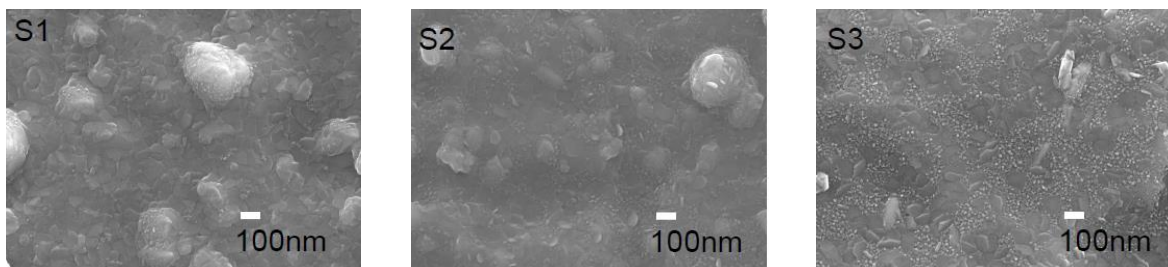


Figure 4. Surface morphologies of the CrAlSiN coatings after TGA/DSC tests at 1000 °C: (a) S1, (b) S2, and (c) S3.

The results of the pin-on-disk tribometer tests for the three CrAlSiN coatings against tungsten carbide ball are shown in Fig. 5. The S1 sample has a lower friction coefficient of 0.5, while the S2 and S3 samples exhibit the slightly high friction coefficient of about 0.5 - 0.55. In addition, the S2 and S3 samples appear to have the more stable sliding curves as compared to the S1 sample. Overall, the effect of the pulsed arc source current on the wear resistance for CrAlSiN coatings against tungsten carbide ball seems to be not remarkable. It is also worth mentioning that all the CrAlSiN coatings obtained in the study have a lower friction coefficient than that of the previous study [6].

Further investigating failure mechanisms, SEM-EDS was used to observe the wear tracks as shown in Fig. 6. The wear track region of the three coatings showed a surface with observable debris agglomerated parallel to the sliding direction on the surface. The S2 and S3 surfaces were severe adhered with debris along the sliding direction, which could be related to the higher friction coefficient. SEM-EDS analysis of the worn track region showed some amounts of oxygen and carbon elements. This infers that the phenomenon of oxidization occurred during the sliding contact, which could be attributed to abrupt temperature rises at the contacting faces during wear test. Since the accumulation and deformation of wear debris happened during the sliding, the wear mechanism of the CrAlSiN coatings against tungsten carbide ball under vertical force caused the adhesive wear.

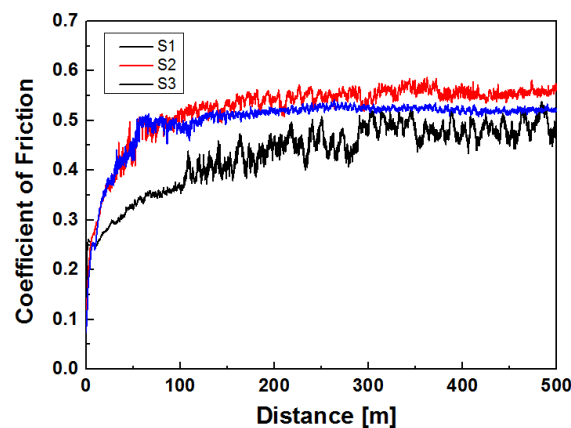


Figure 5. Comparison of the friction coefficient of the three CrAlSiN coatings against a tungsten carbide ball counterface.

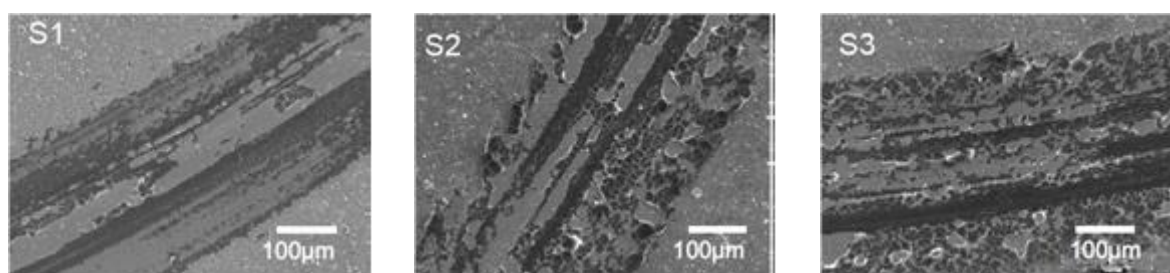


Figure 6. Wear track of the CrAlSiN coatings: (a) S1, (b) S2, and (c) S3.

#### 4. Conclusions

The CrAlSiN coatings were successfully obtained by using pulsed current cathodic arc deposition technique with the Cr and CrAlSi dual targets and N<sub>2</sub> reactive gas. The microstructure analyzed by XRD showed all the coatings are the FCC-Rock salt structure. The CrAlSiN coating deposited with the 90/150 A pulsed current input had the highest hardness value of 4145 HV. After the TGA/DSC analysis in atmosphere, all the coatings were stable up to 1000 °C and the weight ratio was below 0.4.



Pin-on-disk tribometer tests for the CrAlSiN coatings against tungsten carbide ball showed the friction coefficient in the range of 0.4 - 0.5, with the behavior of the adhesive wear in wear mechanism.

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