Effect Of Substrate Surface Roughness On Adhesion Of Titanium Nitride Coatings Deposited By Physical Vapour Deposition Technique

TVSMR Bhushan¹, A Chandrashekhar¹, S Venkat Prasat¹ and I Rajasri Reddy²

¹Department of Mechanical Engineering, Nalla Malla Reddy Engineering College, Hyderabad, India.
²Sumathi Reddy Institute of Technology for Women, Warangal, India.

E-mail-chandubec@gmail.com

Abstract. Medium carbon steel (EN8) substrates with different Surface Roughness (SR) were deposited by Titanium Nitride (TiN). The research work was carried out to investigate the effect of SR and thickness of the thin film coating on substrate structure and tribological properties. EN8 rectangular blocks (60x10x10mm) were used as substrate materials. The variations in SR were achieved by using emery paper on one side and by machining the other side. Scratch tests were conducted and they showed that TiN coatings on substrate with higher SR (1.2 µm) exhibited less adhesion strength compared to TiN coatings on substrate with a lower SR (0.4 µm). Scratch tests also indicated poor adhesion with an increase in the SR of the substrate. The traction force and Coefficient of Friction (COF) were measured during the dry scratch tests under different normal loads, sliding speeds and ramp load conditions. To examine the morphologies of coated substrates and worn surfaces after the scratch test, optical microscopy was utilized. It was found out that TiN coating with a substrate roughness of 0.4 µm exhibited a lower COF and its wear mechanism was adhesive wear. It was experimentally found out that by decreasing the SR of the substrate coatings, wear resistance could be improved.

1. Introduction

Physical Vapour Deposition (PVD) technique is increasingly employed in many engineering applications for protecting the substrate material against wear, corrosion and damage with improvement in the mechanical and tribological properties. PVD techniques have been utilized in automotive industries to reduce the Coefficient of Friction (COF) in sliding interfaces [1]. By depositing the steel substrate materials with coatings like molybdenum disulphide or diamond, researchers have achieved tremendous reductions in wear loss [2].

In recent generation, different types of hard and thin film deposition like PVD techniques and chemical vapor deposition are momentarily engaged. With the help of different thin film depositions such as TiN, AlCrN, TiAlN, CrN and different multi-layer combination of coatings, significant developments in tribological and mechanical properties have been achieved [3]. Totik [4] reported that excellent tribological properties are achieved by the deposition of PVD coatings on steels. In industries, TiN film depositions are trendy and these coatings have showed excellent properties such as low COF, resistance to wear and high hardness. These properties of coatings make them material of choice for
various tribological applications. Bull [5] et al., concluded that "columnar grain are not in close contact in TiN coatings". In mechanical machine parts, moulds, drawing, decorations, bearing and cutting tools, TiN coatings are commonly used [6-7]. During the past two decades many research works have been reported on scratch tests and they were carried out to provide a better understanding of wear mechanisms operating in TiN coatings. Several authors and investigators showed that the volumetric wear loss of steel increases with an increase in the substrate Surface Roughness (SR) of TiN coatings [8-12].

The tribological behaviour of TiN coatings were examined with different coating thickness, substrate hardness, deposition methods and heat treatment. During the frictional contact regime, TiN coating has an affinity to rupture and spall from the substrates. This sort of affinity increases with the rise of applied force, scratch stroke and roughness of substrate materials [13]. In recent years, numerous studies have been conducted on TiN coatings with different substrate roughness. One of the most important characteristics of engineering materials is surface quality. Optimum values of SR provide improvements in the mechanical and tribological characteristics of the machine parts. The SR also plays a key role in economical production and high quality of products. SR also directly affects tool wear.

Scratch tests are widely used to investigate the coating adhesion strength. A scratch test comprises of a moving diamond indenter which introduces stresses by deforming the surface. The applied normal load is increased continuously or incrementally until the coating gets completely detached from the surface. Critical load (Lc) was identified with the help of tractional force values at which the thin film is detached from the substrate material.

In various engineering applications including excavators, mining equipment, pumping part, railways, rolling milling equipments for factories and wear resistant parts of machining elements, steel is used commonly as it has superior qualities in weldability and ductility. Due to the rapid developments in DC PVD techniques, surface pretreatment and preparation of substrate like grinding, sand blasting, polishing and honing are required for better performance of the hard coatings. The effect of substrate SR and thickness of the coatings on deposition of TiN with EN8 substrate has been investigated through experiments with help of micro-scratch test indenter. Investigations were carried out using TiN coatings of 1.5, 3 and 5 µm thickness and the 3 different SR (Ra) values of 0.02, 0.15 and 0.35 µm. The authors reported that Lc decreases with increase in the substrate SR.

One of the most important property in mechanical application is adhesion of coatings on substrate SR. Based on the literature survey it was found out that the effect of substrate SR on TiN deposition has not been discussed methodically. In this research work, a systematic study was conducted on TiN coatings deposited on two medium carbon steel substrates with different SR values by PVD method at Oerlikon Balzers’ Coatings, Pune, India. The main objective of the current work is to predict the effect of different substrate SR structure on TiN coatings. To characterize the mechanical properties of the thin film coatings, scratch test was used.

2. Material and Methods
2.1. Material
TiN of 2 µm coating thickness was coated on 0.436% C medium carbon steel by DC arc PVD method. With the help of "Optical Emission Spectrometer of Thermo Electron S. A. EnVallaire Quest 1024, Ecublens, Switzerland make", the EN8 substrate material’s actual composition has been examined. Table 1 represents the chemical composition of the EN8 steel substrate and sizes of samples 60mm x 25mm x 4mm were cut from the sheet metal. The specimens were initially polished by sandpapers of 150 grade and 1000 grade grit sizes. The specimens were subsequently polished using 1/0 and 4/0 grades on one surface and the other surface was machined.
Table 1. Chemical composition (wt %) of EN8 Steel.

| Elements | C    | Mn   | Si    | P    | S    | Ni   | Cr   | Mo   | Fe   |
|----------|------|------|-------|------|------|------|------|------|------|
| Composition | 0.4360 | 0.7354 | 0.1954 | 0.0230 | 0.0185 | 0.0054 | 0.0115 | 0.0136 | Bal. |

2.2. Deposition Method

The sample used was medium carbon steel substrates, comprising of 0.417%C. To eliminate oxides and other contaminated particles from the EN8 substrate materials, grinding with SiC paper was employed. Cleaned EN8 substrate materials were degreased with acetone and then cleaned with alkali solution containing sodium carbonate and sodium hydroxide at 60°C for 4 minutes of time. These substrate materials were then thoroughly rinsed in distilled water for a minute. Then ~2μm of TiN coating thickness was deposited on EN8 steel. The Oerlikon Balzer machine is provided with 6 cathodic arc sources. 2 of 6 sources were used to deposit a 0.3μm TiN coating thick sub-layer to increase coating adhesion. For depositing the main layer of coatings on substrate materials remaining four sources were used, which was gained by usage of sintered targets. Table - 2 represents the outline of the parameters.

Table 2. Outline of the parameters for TiN reactive sputtering deposition.

| Parameters                              | Values                  |
|-----------------------------------------|-------------------------|
| Target Composition                      | TiN coating: Ti99.9     |
| Bias Voltage (V)                        | -40 V to -170 V         |
| Reactive Gas                            | Nitrogen                |
| Substrate                               | 0.4%C                   |
| Target to substrate distance, mm        | 60                      |
| Substrate temperature, C                | 450°C ± 10°C            |
| Coating Thickness                       | 2 μm ± 1 μm             |

2.3. Microstructure

Metallurgical optical microscope ("Zeiss Axiovert 200 MAT inverted optical microscope") experiments were carried out to analyze the microstructures of coated substrate materials before and after the scratch test. Version 4.1 Zeiss Axiovision image analyzer was used to analyze the microstructure. Based on the ASTM Standard B276, the measurements of porosity were made. Researchers performed the examination with help of Dewinter Materials Plus 1.01 software image analyzer and acquired the microstructure images of coated EN8 substrate materials by using a PMP3 inverted metallurgical microscope.

2.4. Scratch test

The DUCOM scratch tester TR-101 apparatus used in this present work is shown in figure 1. The stylus Rockwell C diamond indenter has 0.04cm end tip diameter. The researchers scratched the spherical tip stylus across the coatings at the normal load rate of 10 N/mm and scratch stroke distance of 1 mm/sec. To determine the results during the start of the scratch track, an applied force of 10N was applied and at 14mm stroke length the scratch test was stopped. Normal force, tractional force and COF readings were recorded in computer during the experiments. The number of failure events which occurred along with the scratch track were analyzed under different operational conditions.
3. Experimental results and discussions

3.1. Microstructure analysis
The TiN coatings were deposited successfully by PAPVD (Plasma assisted physical vapour deposition) processes on EN8 steel. The optical micrographs of the substrate and TiN coatings are depicted in Figure 2. Figure 2 (a) shows the microstructure of EN8 steel; the lighter portion is pearlite and the darker portion is ferrite. The surface appearance of substrate is 50% ferrite structure and 50% pearlite structure, whereas in case of TiN (Figure 2b) coating is gold-yellow in color. Deposited coatings have uniform microstructure and obviously it contains some inclusions and pores. The porosity for as coated TiN coating is 0.52.

3.2. Scratch test analysis
Thin film scratch tester was used for measurement of adhesion strength. The normal load which was applied on TiN coatings was progressively increased along the scratch track. The increase in the applied normal load with the variations in the stroke length is shown in Figures. 3(a) and 3(b). The traction force and COF were measured when the normal loads were increased. Figures. 3(a) and 3(b) exhibit the variations of the applied normal loads on TiN coating with substrates of both lower and
higher SR for various sliding speeds. The lowest and highest loads at which the coating damage occurred was determined by measuring the traction force and COF. All the tests were repeated more than two times in order to get the exact values of COF [14-16].

Figures 3(a) and 3(b) show the values of the normal load increasing along the stroke length. The normal load diagram indicates that there was a reduction in applied force at the end of the stroke length, especially for 10 N/mm rate of loading. This phenomenon can be attributed to the complete removal of the coating from the sample and this indicated a failure due to adhesive wear mechanism.

The scratch test results indicating the values of traction force at various stroke lengths are illustrated in Figure 4. For the medium carbon steel substrate, tractional load raised progressively with a rise in the scratch stroke. The step-like behavior of the load variation and scratch track obtained on coating of 2N/mm rate of loading indicates the gradual appearance of the coating defects. The shape of the curve obtained for 5 N/mm rate of loading is wavy in nature and there is only a slight change in slope of the curve when compared to the other curves. The plot obtained for the traction force vs stroke length at 10 N/mm rate of loading indicates numerous irregularities, which depicts the occurrence of severe wear mechanism and failure of the coating as well as the substrate surface [17-19]. The waviness in the shape of the curves decreased with increase in the indenter normal loads. The greater slope of the traction force curve indicates that the share of elastic deformation increases with an increase in stylus depth.

Figure 5 indicates the changes of COF with rise in the normal force applied on the TiN. The values of COF obtained for substrate with SR 0.4 µm was lower in comparison to the COF obtained for substrate of SR 1.2 µm. This phenomenon could be adhesion characterization and cohesion of the film with the substrate. It can be inferred from the graph that chipping and flaking simply arises at an initial phase of the experiments in the coating with higher SR (1.2 µm). The coated wear debris particles also piled up right in front of the stylus indenter, which hampered the free movement of the indenter and this increased the COF. From Figure 5(a), it was noticed that at the initial stage COF increases and then decreases suddenly at 0.32 COF value with respect to 43 N, which may be due to cohesive failure in the coating. At the starting applied force of 40 N, scratch speed of 0.1 mm/sec and rate of loading of 10 N/mm, the test results indicate tremendous variation at the end of the scratch stroke, the reason may be due to severe plastic deformation and adhesive failure (Fig. 5b). Therefore, superior adhesion and cohesion of the TiN coatings with the smoother substrate SR showed lower COF results [20-21].
Figure 4. Difference of tractional load vs scratch stroke of the TiN coating on EN8 substrate sample (0.4 µm & 1.2 µm) with different sliding speeds as recorded during the scratch test.

Figures 5 (a) and (b). Variation of COF vs normal load for the TiN coating on EN8 substrate sample (a) start load 10N and (b) start load 40 N with respective sliding speeds.

Figure 6 depict that progressively the COF was increased with the scratch stroke distance as the normal load was increased and the rupture mode transformed from cohesion to adhesion. It was also observed that lower COF values were obtained for the substrate with lower SR [22-23]. The present investigation shows that the coating adhesion strength increases with a decrease in the SR of the materials. The coating adhesion obtained on lower SR substrate (0.4 µm) was superior to coating adhesion obtained on higher SR substrate (1.2 µm).
4. Conclusions

Materials with TiN coatings and different SR have been systematically examined. The present investigation demonstrated that the thin film coating bonding strength increases with a decrease in the SR of materials. The coating adhesion obtained on lower SR substrate (0.4 µm) was superior to coating adhesion obtained on higher SR substrate (1.2 µm). It was found out that TiN coating with a substrate roughness of 0.4 µm exhibited a lower COF and its wear mechanism was adhesive wear. It was experimentally found out that the coatings wear resistance could be improved by decreasing the SR of the materials.

5. References

[1] Chawla Vikas, Puri D, Prakash S, Chawla Amit and Sidhu Singh Buta(2009) Characterization and Comparison of Corrosion Behavior of Nanostructured TiAlN and AlCrN Coatings on Superfer 800H (INCOLOY 800 H) Substrate Journal of Minerals & Materials Characterization & Engineering, 8715-727

[2] Subramanian B, Ashok K and Jayachandran M, (2008) Effect of substrate temperature on the structural properties of magnetron sputtered titanium nitride thin films with brush plated nickel interlayer on mild steel Applied Surface Science, 255 2133-2138

[3] Subramanian B and Jayachandran M,(2007) Characterization of reactive magnetron sputtered nanocrystalline titanium nitride (TiN) thin films with brush plated Ni interlayer J ApplElectrochem, 37,1069-1075

[4] Valli J and Makela U, Matthews A and Murawa(1985)TiN coating adhesion studies using the scratch test method J Vac Sci Technol, 3, 2411-2414

[5] Takadoum J and Bennani Houmid H (1997) Influence of substrate roughness and coating thickness on adhesion, friction and wear of TiN films Surface and Coatings Technology96272-282

[6] Stallard J, Poulat S and Teer D G, (2006) The study of the adhesion of a TiN coating on steel and titanium alloy substrates using a multi-mode scratch tester Tribology International 39159–166

[7] Ozek, Cebeli, Taşdemir andVedat, (2009) Modeling of surface roughness with variance analysis in turning of AISI 304 austenitic stainless steel e-Journal of New World Sciences Academy, 4,305
[8] Huang Ruo-xuan, Qi Zheng-bing, Sun Peng, Wang Zhou-cheng and Wu Chong-hu (2011) Influence of substrate roughness on structure and mechanical property of TiAlN coating fabricated by cathodic arc evaporation Physics Procedia, 18:160-167

[9] Yana Rüdenauer (2013) Influence of Interfacial Parameters on the Adhesion of Soft Polymers Universität Karlsruhe

[10] Narasimman P, Pushpavanam Malathy and Periasamy V M, (2012) Wear and scratch resistance characteristics of electrodeposited nickel-nano and micro SiC composites Wear, 292:293,197-206

[11] Seo Tae-Won and Weon Jong-Il (2012) Influence of weathering and substrate roughness on the interfacial adhesion of acrylic coating based on an increasing load scratch test J Mater Sci, 47:2234-2240

[12] Bhaduri D and Chattopadhyay A K (2010) Study on the role of PVD TiN coating in improving the performance of electroplated monolayer superabrasive wheel Surface & Coatings Technology 205 658-667

[13] He Y, Apachitei I, Zhou J, Walstack T and Duszczyk J (2007) The influence of the depth of a plasma nitrided layer in tool-steel substrate on the scratch-resistant properties of PACVD TiBN coating Surface & Coatings Technology 201:7036–7042

[14] Subramanian B, Ashok K, Subramanian K, Sastikumar D and Selvan G, Jayachandran M, (2009) Evaluation of corrosion and wear resistance titanium nitride (TiN) coated on mild steel (MS) with brush plated nickel interlayer Surface Engineering, 25, 490-495

[15] Chawla Vikas, Puri D, Prakash S, Chawla Amita and Sidhu Singh Buta (2009) Characterization and Comparison of Corrosion Behavior of Nanostructured TiAlN and AlCrN Coatings on Superfer 800H (INCOLOY 800 H) Substrate Journal of Minerals & Materials Characterization & Engineering, 8:715-727

[16] Subramanian B, Ashok K and Jayachandran M, (2008) Effect of substrate temperature on the structural properties of magnetron sputtered titanium nitride thin films with brush plated nickel interlayer on mild steel Applied Surface Science, 255, 2133-2138

[17] Subramanian B and Jayachandran M.(2007) Characterization of reactive magnetron sputtered nanocrystalline titanium nitride (TiN) thin films with brush plated Ni interlayer J Appl Electrochem, 37, 1069-1075

[18] Valli J and Makela U, Matthews A and Murawa (1985)TiN coating adhesion studies using the scratch test method J Vac Sci Technol, 3,2411-2414

[19] Takadoum J and Bennani Houmid H (1997) Influence of substrate roughness and coating thickness on adhesion, friction and wear of TiN films Surface and Coatings Technology 96:272-282

[20] Stollard J, Poulat S and Teer D G, (2006) The study of the adhesion of a TiN coating on steel and titanium alloy substrates using a multi-mode scratch tester Tribology International 39:159–166

[21] Ozek, Cebeli, Taşdemir and Vedat, (2009) Modeling of SR with variance analysis in turning of AISI 304 austenitic stainless steel e-Journal of New World Sciences Academy, 4,305

[22] Huang Ruo-xuan, Qi Zheng-bing, Sun Peng, Wang Zhou-cheng and Wu Chong-hu (2011) Influence of substrate roughness on structure and mechanical property of TiAlN coating fabricated by cathodic arc evaporation Physics Procedia, 18,160-167

[23] Yana Rüdenauer, (2013) Influence of Interfacial Parameters on the Adhesion of Soft Polymers Universität Karlsruhe

[24] Narasimman P, Pushpavanam Malathy and Periasamy V M, (2012) Wear and scratch resistance characteristics of electrodeposited nickel-nano and micro SiC composites Wear, 292:293,197-206

[25] Seo Tae-Won and Weon Jong-Il (2012) Influence of weathering and substrate roughness on the interfacial adhesion of acrylic coating based on an increasing load scratch test J Mater Sci, 47:2234-2240
[26] Bhaduri D and Chattopadhyay A K, (2010) Study on the role of PVD TiN coating in improving the performance of electroplated monolayer superabrasive wheel Surface & Coatings Technology 205 658-667

[27] He Y, Apachitei I, Zhou J, Walstock T and Duszczyk J (2007) The influence of the depth of a plasma nitrided layer in tool-steel substrate on the scratch-resistant properties of PACVD TiBN coating Surface & Coatings Technology 201 7036–7042

[28] Subramanian B, Ashok K, Subramanian K, Sastikumar D, Selvan Gand Jayachandran M, (2009) Evaluation of corrosion and wear resistance titanium nitride (TiN) coated on mild steel (MS) with brush plated nickel interlayer Surface Engineering, 25 490-495