Optimization and Characterization of Si$_3$N$_4$ Layer for Wear Resistant Ti-Al-N/Si$_3$N$_4$ Nano-Composite Coatings

Balaji N, Sivakumar C, Arun kumar P

Abstract—Machining Custom 465 steel at high speeds usually at 20-30% lower than ordinary stainless steel leads to excessive tool wear and thereby affecting the tool life. In such case tool life can be increased by coating a nano-composite layer of Ti-Al-N/Si$_3$N$_4$ which exhibits high strength, hardness, toughness, resistance to oxidation and thermal shock. Particularly, Si$_3$N$_4$ serves as an interfacial phase in nano-composite layer is thermodynamically stable phase at high temperatures up to 1850°C along with high oxidation resistance which might reduce the heat flow between tool-workpiece interfaces leading to high thermal stability of cutting tool. Physical vapor deposition techniques can be used to develop Ti-Al-N/Si$_3$N$_4$ nano-composite coating which involves typically 81 trial depositions in order to optimize process parameters for Si$_3$N$_4$. Here we attempt to reduce the number of trials by design and optimization of process parameters which was efficiently achieved at faster rate by applying the Taguchi design. In present work, we used Taguchi orthogonal (L$_9$) array to conduct the 9 experiments and obtained optimum process parameters for Si$_3$N$_4$ coating. Based on the design we deposited Si$_3$N$_4$ nanocoating using RF magnetron sputtering process with 4 factor and 3 level process parameters namely, Ar/N$_2$ gas mixture, RF Power, deposition time, and deposition pressure on high speed steel (HSS), tungsten carbide (WC) and Si (100) substrates. Atomic Force Microscopy (AFM) studies were carried for surface roughness, topography, and phase contrast imaging. Glancing incidence X-ray diffraction (GIXRD) studies were performed for the identification and quantification of crystalline and amorphous phase. Field Emission Scanning Electron Microscopy (FE-SEM) studies were performed for the film thickness and grain size of Si$_3$N$_4$ layers.

Keywords: Wear, Nano-composite, Optimization, X-ray diffraction, Sputtering

I. INTRODUCTION

Machining a harden material like Custom 465 steel in dry condition without using coolant is a greater challenge of advance manufacturing because of its existing a natural properties, like low density and have low thermal conductivity, low modulus of elasticity and high chemical reactivity with other materials at elevated temperature. In high grade material using lubricant and coolant it react with material composition and it may occur corrosion and toxic. To avoid this industries may go for dry machining process in future, it will act as a part of ecological protection. In industries process they can reduce machining cost instead of coolant and lubricant.It will reduce 15% of total cost in account of wet machining.

Revised Manuscript Received on July 05, 2019.

Balaji N, Department of Mechanical Engineering, Vel Tech Rangarajan Dr. Sagunthala R&D Institute of Science Technology, Tamil Nadu, India.

Sivakumar C, Department of Mechanical Engineering, Vel Tech Rangarajan Dr. Sagunthala R&D Institute of Science Technology, Tamil Nadu, India.

Arun kumar P, Department of Mechanical Engineering, Vel Tech Rangarajan Dr. Sagunthala R&D Institute of Science Technology, Tamil Nadu, India.
II. EXPERIMENTAL DETAILS

Deposition of coating is coated with a standard DANVEC glovebox PVD coating machine. The construction of machine attached both RF and DC sputtering/pulsed DC sputtering with 3” target with co-sputtering capability. Target of $\text{Si}_3\text{N}_4$ is 99% purity with Copper backing plate, used to coat a 100 nm thick of $\text{Si}_3\text{N}_4$ layer on the high-speed steel (HSS), tungsten carbide (WC) and Si (100) substrates. The high speed steel (HSS), tungsten carbide (WC) and Si (100) substrates materials are prepared from their parent materials which is of size 1 x 1 inch. The target in shape of Circular and the material is sintered in circular disc, during the coating process it will placed in vacuum chamber at the time of coating.

Prior to deposition, for Silicon and HSS coating materials were carrying four step of cleaning process like — first with an Acetone bath for 5 min in ultrasonic device, second with an Isopropyl alcohol ultrasonic bath for 5 min, third with an DI–Ionized water ultrasonic bath for 5 min and finally with an HF with 2% DI water ultrasonic bath for 5 min. Then dried under room temperature.

Tungsten carbide substrates are cleaned using Acetone bath for 5 min. A line diagram of the PVD chamber is display in Fig. 1. For $\text{Si}_3\text{N}_4$ coatings the target material was placed in Gun stand were inside of vacuum chamber, then target material will deposit on all the three materials, are high speed steel (HSS), tungsten carbide (WC) and Si (100) substrate.

Nano-composite coating which involves typically 81 trial depositions in order to obtain optimized process parameters for Si3N4. Here we attempt to reduce the number of trials by design and optimization of process parameters which was efficiently achieved at faster rate by applying the Taguchi design. Taguchi orthogonal (L9) array as shown in Table 2. was employed to conduct the 9 experiments and to obtain optimum process parameters for $\text{Si}_3\text{N}_4$ coating. Based on the design we deposited $\text{Si}_3\text{N}_4$ nanocoating using RF magnetron sputtering process with 4 factor and 3 level process parameters namely, Ar: $\text{N}_2$ gas mixture, RF Power, deposition time, and deposition pressure. The Ar: $\text{N}_2$ reactive gas mixture added during the evaporation, with trail value flow rate. Then the ionization and excitation occur during the interaction with silicon target with the reaction of plasma. Then the ionized metal which form very hard thin films on the substrate surface. The PVD plasma is protected in vacuum chamber by passing Ar (argon gas) inside the vacuum chamber. The reactive and inert gas maintained by chamber pressure gauge. Table 3. shows the detail actual readings obtained during the trial experiments.

![Figure 1. Line diagram of the PVD deposition system](image)

### Table 2. Design of Experiments for $\text{Si}_3\text{N}_4$

| Sample ID | Ar:$\text{N}_2$ Gas Flow Rate (SCCM) | RF Power (W) | Operating Pressure (mbar) | Deposition Time (Min) |
|-----------|-------------------------------------|--------------|---------------------------|-----------------------|
| SN1       | 5:5                                 | 100          | $1.52 \times 10^{-2}$     | 10                    |
| SN2       | 5:5                                 | 200          | $2 \times 10^{-2}$        | 20                    |
| SN3       | 5:5                                 | 300          | $2.5 \times 10^{-2}$      | 30                    |
| SN4       | 5:7.5                               | 100          | $2 \times 10^{-2}$        |                        |
Table 3. Actual Readings during experiments

| S.No | Sample ID | Base Pressure (mbar) | Ar:N₂ Gas Flow Rate (SCCM) | RF Power (W) | Operating Pressure (mbar) | Temp (°C) | Rotation (RPM) | Deposition Time (Min) |
|------|-----------|----------------------|-----------------------------|--------------|---------------------------|-----------|----------------|----------------------|
| 1    | SN1       | 1.20 x 10⁻⁶          | 5:5                         | 100          | 1.52 x 10⁻²               | 300       | 20             | 10                   |
| 2    | SN2       | 2.79 x 10⁻⁶          | 5:5                         | 200          | 2 x 10⁻²                  | 300       | 20             | 20                   |
| 3    | SN3       | 3.5 x 10⁻⁶           | 5:5                         | 300          | 2.51 x 10⁻²               | 300       | 20             | 30                   |
| 4    | SN4       | 2.53 x 10⁻⁶          | 5:7.5                       | 100          | 2 x 10⁻²                  | 300       | 20             | 30                   |
| 5    | SN5       | 3 x 10⁻⁶             | 5:7.5                       | 200          | 2.5 x 10⁻²                | 300       | 20             | 10                   |
| 6    | SN6       | 1.86 x 10⁻⁶          | 5:7.5                       | 300          | 1.52 x 10⁻²               | 300       | 20             | 20                   |
| 7    | SN7       | 2 x 10⁻⁶             | 5:10                        | 100          | 2.5 x 10⁻²                | 300       | 20             | 20                   |
| 8    | SN8       | 2 x 10⁻⁶             | 5:10                        | 200          | 1.6 x 10⁻²                | 300       | 20             | 30                   |
| 9    | SN9       | 2 x 10⁻⁶             | 5:10                        | 300          | 2.01 x 10⁻²               | 300       | 20             | 10                   |

The coated samples are characterized done with Atomic Force Microscopy (AFM) for 3D Surface topology, Field Emission Scanning Electron Microscope (FE-SEM) for coating microstructure and cross sectional of coating thickness, and chemical composition analyzed with EDS.

III. Results and Discussion

Microstructure Analysis for Coating

The top layer coated with Si₃N₄ characterization images are arranged in trail order as per L9 design of experiment. The coated substrates are characterized in different levels depend upon the trail method. Each and every trail will differ with given parameters, some substrates coated will, another substrates coated unevenly and some substrates are not coated depends on current levels.

In Field emission scanning electron microscope (FE-SEM) images are clear to see the coating on substrates, and also good bonded with substrates material. In microstructure image number SN2, SN6, SN7 and SN8 are coated uniformly and less pores on coated layer. Image number SN1 and SN3 not coated will in substrates, trail variations of parameter value (Current value) will be minimum, for deposition current value not sufficient and coating time also very low.

The SEM images are nine trial coatings in glass substrates. In Si₃N₄ deposition process, silicon and nitrogen deposit perfectly in substrates. Coatings show a fine and smooth morphology in surface. Some hairline crack presence in substrate due to Nitrogen gas deposition. But hairline cracks do not promote during machining process and it also called as gargling effect. Some microporous also visible in Si₃N₄ coated surfaces. After the deposition process the micro particle will drop out from substrates.
Optimization and Characterization of Si₃N₄ Layer for Wear Resistant Ti-Al-N/Si₃N₄ Nano-Composite Coatings

Figure 2. Microstructure of the Si₃N₄ coating

Figure 3. Cross section of the Si₃N₄ coating
Table 4 Elemental composition of the $\text{Si}_3\text{N}_4$

| S.No | Sample ID | Silicon % | Nitrogen % |
|------|-----------|-----------|------------|
| 1    | SN1       | 96.39     | 3.61       |
| 2    | SN2       | 98.51     | 1.49       |
| 3    | SN3       | 94.61     | 5.39       |
| 4    | SN4       | 98.65     | 1.35       |
| 5    | SN6       | 94.79     | 5.21       |
| 6    | SN7       | 98.67     | 1.33       |
| 7    | SN8       | 96.71     | 3.29       |
| 8    | SN9       | 98.67     | 1.33       |

Figure 4 EDS chemical composition graph and phase contrast
IV. SURFACE MORPHOLOGY

The surface morphology scanned using Atomic Force Microscopic (AFM). In scanning process 0-10 µm range set as maximum range for scanning, and the maximum probe moving speed in substrate is 0.1 mm/s. In SN2 substrates shows irregularities due to some cracks or corrosion in substrates. Which can be poor surface roughness, the rough surface will wear and tear quickly and also have high friction coefficient than the SN1 substrate. The waviness of peak and valley shows irregularities in deposition, the grains are disorientation in surface. The SN trail substrates are coated with mixed grains silicon and Nitrate (Si₃N₄). The AFM results shows in Table 6. Figure 6 shows the 3D AFM images of the Si₃N₄coating.

| S.No | Sample ID | Thickness (nm) |
|------|-----------|----------------|
| 1    | SN1       | 81.2           |
| 2    | SN2       | 98.4           |
| 3    | SN3       | 103.6          |
| 4    | SN4       | 45.68          |
| 5    | SN6       | 98.46          |
| 6    | SN7       | 62.5           |
| 7    | SN8       | 101.2          |
| 8    | SN9       | 75.8           |

Table 6. Surface Roughness for Coated Si₃N₄ Substrate

| S.No | Sample ID | Surface Roughness [nm] |
|------|-----------|------------------------|
| 1    | SN1       | 1.54692                |
| 2    | SN2       | 2.52180                |
| 3    | SN3       | 1.08666                |
| 4    | SN4       | 2.94149                |
| 5    | SN6       | 1.55059                |
| 6    | SN7       | 1.92102                |
| 7    | SN8       | 1.49209                |
| 8    | SN9       | 2.90802                |
Figure 5 AFM 3D Topology and Phase Contrast

| Sample ID | 3D-Topography | 3D-Phase contrast |
|-----------|---------------|-------------------|
| SN1       | ![3D-Topography](image1.png) | ![3D-Phase contrast](image2.png) |
| SN2       | ![3D-Topography](image3.png) | ![3D-Phase contrast](image4.png) |
Optimization and Characterization of Si₃N₄ Layer for Wear Resistant Ti-Al-N/Si₃N₄ Nano-Composite Coatings
SN6

SN7
Optimization and Characterization of Si₃N₄ Layer for Wear Resistant Ti-Al-N/Si₃N₄ Nano-Composite Coatings
V. S/N Ratio for Thickness and Surface Roughness

The optimum combination parameters for producing high thickness and uniform deposition of Si₃N₄ layer were determined as deposition time 20 min, Ar:N₂ flow rate of 5:7.5 sccm, RF sputtering power of 200W, base pressure of 10mTorr.

VI. CONCLUSIONS

The silicon Nitrate (Si₃N₄) coating was deposited on three model (Tungsten, HSS and Silicon substrates and characterization (AFM, SEM and EDS) also done for the trial substrates. The following conclusions are drawn from the characterization results:

1. The SN3 sample coated perfectly with smooth surface on substrate. EDS characterization also confirmed the formation of the composition of the Si₃N₄ depositions. SN3 substrates has 5.39% of Nitrogen content and 94.61% of Silicon content of Which makes the deposition perfect one. These trail deposition is optimized one.

2. The SN3 SEM characterization the micrographs shows that coating deposition is very clear dense, smooth, and has a uniform structure on surface.

3. From SEM cross section characterization, the thickness of coating is desirable and evenly coated with 103.6 nm is obtained.

4. In SN3 substrate AFM scanning images show that the deposition of silicon nitrate coated uniformly, contain less pores and irregularities.

5. All the nine coated substrates were characterized with AFM for 3D Topology and morphology, SEM cross sectional for coating thickness and EDS material composition and mechanical properties. Then the SN3 concluded uniform coated, desirable thickness 103.6 nm, and higher hardness. In AFM scanning image in 3D topology the amorphous Si₃N₄ matrix formed with a strong interfacial bond crystalline phase formation. That so the surface have clear and smooth texture with good roughness value.

6. In Taguchi L9 array is used conduct experiment in nine trial and MINITAB software used to find optimum parameter with S/N Ratio graph with Surface roughness and coating thickness. Optimum parameter for coating thickness take larger is better point, for surface roughness take lower is better optimum parameter. From the optimum parameter SN3 we got smooth surface finish and uniform coating.

REFERENCES

1. Sreejith P S, Ngoi B K A, Dry Machining: Machining of the future, Journal of Materials Processing Technology, Elsevier, 2000, 287 – 291.
2. K. Zhang, L.S. Wang, G.H. Yue, Y.Z. Chan, D.L. Peng.
3. Z.B. Qi, Z.C. Wang, Structure and mechanical properties of TiAlSiN/Si₃N₄ multilayer coating, surface and coating tech. 205(2011)3588-3595.
4. Harish C, Barshilia, B. Deepathi, K.S. Rajam, Deposition and characterization of CrN/Si₃N₄ and CrAlN/Si₃N₄ nano composite coating prepared using reactive DC unbalanced magnetron sputtering, surface and coating tech.201(2007)9468-9475.
5. Yanqiu Xiu, Shinya Sasaki, Takashmi Murakami, Miki Nakano, Leishi Haizhong Wang, Ionic liquid lubrication of electrodeposited nickel-si₃n₄ composite coating, wear262(2007)765-771.
6. E. Uhlmam, J.A. Oyanedel, R. GershenBerger, H. Frank, nc-AlTiN/a-si₃n₄ and nc-AlCrN/a-si₃n₄ nano composite coating as protection layer for PCBN tools in Hard Maching, surface and coating tech. 237(2013)142-148.
Optimization and Characterization of Si$_3$N$_4$ Layer for Wear Resistant Ti-Al-N/Si$_3$N$_4$ Nano-Composite Coatings

7. Zhiqiang Liu, Qingiong An, Jinyang Xu, Ming Chen, Shu Han. Wear performance of (nc-AlTiN)/a- Si$_3$N$_4$ and (nc-AlCrN)/a- Si$_3$N$_4$ coating in high speed machining of titanium alloy under dry and minimum quantity lubrication (MQL) condition, wear 305(2013)249-259.

8. Donghai Yu, Chengyong Wang, Xiaoling Cheng, Fenglin Zhang. Optimization of hybrid PVD process of TiAlN Coating by Taguchi method, applied surface science 255(2008)1865-1869.

9. A.R. Bushroa, H.H. Masjuki, M.R. Muhamad. Parameter optimization of sputtered Ti interlayer using taguchi method, IJIME, vol.6 (2011), no.2.140-146.

10. Ray Kundaliya, D.P. Shanubhogue. A. Comparison study; Taguchi methodology through a case study of accelerated failure in spin-on-filter, engineering and technology vol.2, Issue3, March 2015.

11. Prabakaran M, Suresh Kumar S, Ramyesh K.R, Srinivasan R.V. Characterization and optimization of CrN coating on tool steel (6959), IJMIT vol.2, Issue1, pp: (108-112) 2014.

12. Aman Aggarwal, Hari Sing, Pradeep Kumar, Manmohan Sing. Optimizing power consumption for CNC turned parts using response surface methodology and taguchi technique-A comparative analysis material processing technology 200(2008)373-384.

13. Mohd Sazali Md Said, Jaharah A. Ghanı, MohdShahir Kassim, Siti Haryani Tomadi, Che Hassan Che Haran. Comparison between Taguchi method and response surface methodology (RSM) in optimizing Machining Condition, Robust quality engineering.

14. V. Ezhil Selvi, V.K. William Grips, Harish C. Barshilia. Electro chemical behavior of super hard nano composite coating of TiN/si3n4 prepared by reactive DC unbalanced Magnetron sputtering. Surface and coating technology 224(2013)42-48.

AUTHORS PROFILE

N Balaji, Assistant Professor, Vel Tech Rangarajan Dr Sagunthala R&D Institute Of Science And Technology, Nbalaji@Veltech.Edu.In
Research In Nano Coating Composite Layer

P Arun Kumar, Assistant Professor, Vel Tech Rangarajan Dr Sagunthala R&D Institute Of Science And Technology, Arunkumarpp@Veltech.Edu.In

C Sivakumar, Assistant Professor, Vel Tech Rangarajan Dr Sagunthala R&D Institute Of Science And Technology, sivakumarc@veltech.edu.in