Optimization and Characterization of Si_3N_4 Layer for Wear Resistant Ti-Al-N/Si_3N_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_3N_4 which exhibits high strength, hardness, toughness, resistance to oxidation and thermal shock. Particularly, Si_3N_4serves 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_3N_4 nano-composite coating which involves typically 81 trial depositions in order to obtain optimized process parameters for Si_3N_4. Here we attempt to reduce the number of trails 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 (L9) array to conduct the 9 experiments and obtained optimum process parameters for Si3N4 coating. Based on the design we deposited Si3N4nanocoating using RF magnetron sputtering process with 4 factor and 3 level process parameters namely, Ar:N2 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 Si3N4layers.

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.

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So in order to machine such hard materials under dry environment the cutting tool material should meet the specific requirements such as high hardness in combination with superior thermal and chemical stability, corrosion and high temperature oxidation resistance. To withstand high thermal conductivity and chemical stability, the material properties are more important to improve the tool performance.Peculiarly in machining process loads act on the cutting edges, like thermal and mechanical load.

This can be achieved by coating a nano-composite layer of Ti-Al-N/Si_3N_4 over cutting tool material which exhibits high strength, hardness, toughness, resistance to oxidation and thermal shock. Particularly, Si_3N_4serves as an interfacial phase in nano-composite layeris thermodynamically stable phase at high temperatures up to 1850˚C along with high oxidation resistance which might reduce the heat flowbetween tool- workpiece interfaces leading to high thermal stability of cutting tool.

Titanium Nitride (TiN) coatings is widely used to improve the properties of cutting tools in terms of their life and functionality. But TiN coatings readily oxidize at 500^{o}C, limiting their applicability in high speed and dry machining. Addition of Al in the TiN phase leads to improvement in oxidation and wear resistance upto 800^{o}C. However when the fine grain crystals of TiAlN or AlTiN phase are embedded in amorphous silicon nitride matrix,then Ti-Al-N /Si_3N_4 coating shows an improved hardness and oxidation resistance at high temperatures of more than 1200^{o}C. These coatings show very apt properties to perform dry machining under high speed conditions [2 – 6].

Various techniques are available to deposit the thin films on cutting tools. The Chemical Vapor Deposition (CVD) and Physical Vapor Deposition (PVD) techniques are two broad techniques. CVD techniques possess certain problems such asthe high temperature during deposition which is nearly 1000^{o}C. Introducing ammonia gas instead of the nitrogen increases concentration of chlorine and causes flaking of coatings. The PVD coating technology is the most promising technique widely used in industries today.

Physical vapor deposition techniques has be used to develop Ti-Al-N/Si_3N_4 nano-composite coating which involves typically 81 trial depositions in order to obtain optimized process parameters for Si_3N_4. Here we attempt to reduce the number of trails 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 experiment sand obtained optimum process parameters for Si_3N_4coating. The aim of present work involves indesigning, optimization and processing of Si_3N_4 layer for wear resistant Ti-Al-N/Si3N4 nano- composite coatings.

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 Si_3N_4 is 99% purity with Copper backing plate,used to coat a 100 nm thick of Si_3N_4layer on thehigh 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 Si_3N_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.

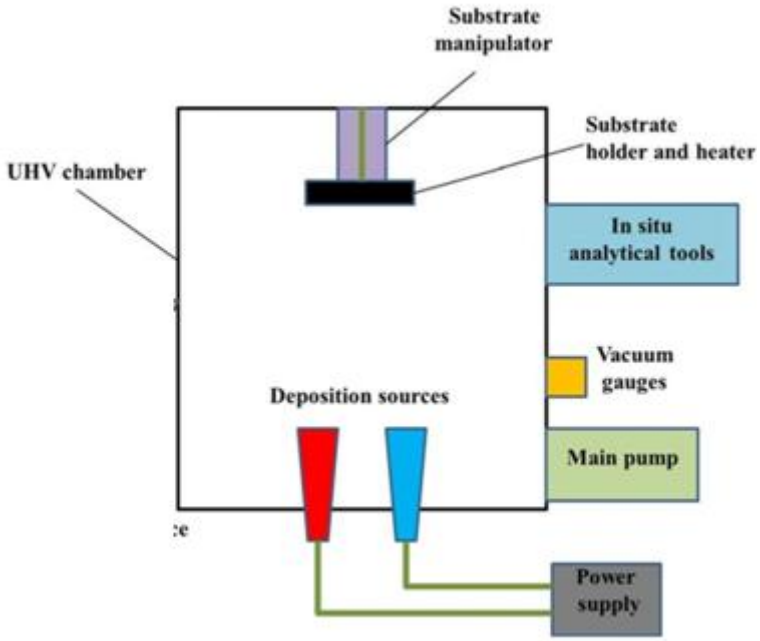


Figure 1. Line diagram of the PVD deposition system

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 trails 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 Si_3N_4 coating. Based on the design we deposited Si_3N_4nanocoating 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. The Ar: 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 trail experiments.

Table 2. Design of Experiments for Si_3N_4

Sample ID	Ar:N_2 Gas Flow Rate (SCCM)	RF Power (W)	Operating Pressure (mbar)	Deposition Time (Min)
SN1	5:5	100	1.52×10^{-2}	10
SN2	5:5	200	2×10^{-2}	20
SN3	5:5	300	2.5×10^{-2}	30
SN4	5:7.5	100	2×10^{-2}	30

SN5	5:7.5	200	2.5×10^{-2}	10
SN6	5:7.5	300	1.52×10^{-2}	20
SN7	5:10	100	2.5×10^{-2}	20
SN8	5:10	200	1.52×10^{-2}	30
SN9	5:10	300	2×10^{-2}	10

Table 3. Actual Readings during experiments

S.No	Sample ID	Base Pressure (mbar)	Ar:N_2Gas Flow Rate (SCCM)	RF Power (W)	Operating Pressure (mbar)	Temp (C°)	Rotation (RPM)	Deposition Time (Min)
1	SN1	1.20×10^{-6}	5:5	100	1.52×10^{-2}	300	20	10
2	SN2	2.79×10^{-6}	5:5	200	2×10^{-2}	300	20	20
3	SN3	3.5×10^{-6}	5:5	300	2.51×10^{-2}	300	20	30
4	SN4	2.53×10^{-6}	5:7.5	100	2×10^{-2}	300	20	30
5	SN5	3×10^{-6}	5:7.5	200	2.5×10^{-2}	300	20	10
6	SN6	$1.86x\ 10^{-6}$	5:7.5	300	1.52×10^{-2}	300	20	20
7	SN7	2×10^{-6}	5:10	100	2.5×10^{-2}	300	20	20
8	SN8	2×10^{-6}	5:10	200	$1.6x\ 10^{-2}$	300	20	30
9	SN9	2×10^{-6}	5:10	300	2.01×10^{-2}	300	20	10

The coated samples are characterization 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

MicrostructureAnalysis for Coating

The top layer coated with Si_3N_4characterization images are arranged in trail order as per L9 design of experiment. The coated substrates are in different levels depend upon the trail method. Each and every trail will differs 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 Si_3N_4 deposition process, silicon and nitrogen deposit perfectly in substrates. Coatings show a fine and smooth morphology in surface. Some hair line crack presence in substrate due to Nitrogen gas deposition. But hair line cracks do not promote during machining process and it also called as garglingeffect.Some microporousalsovisible in Si_3N_4 coated surfaces. After the deposition process the micro particle will drop out from substrates.

Figure 2. Microstructure of the Si_3N_4 coating

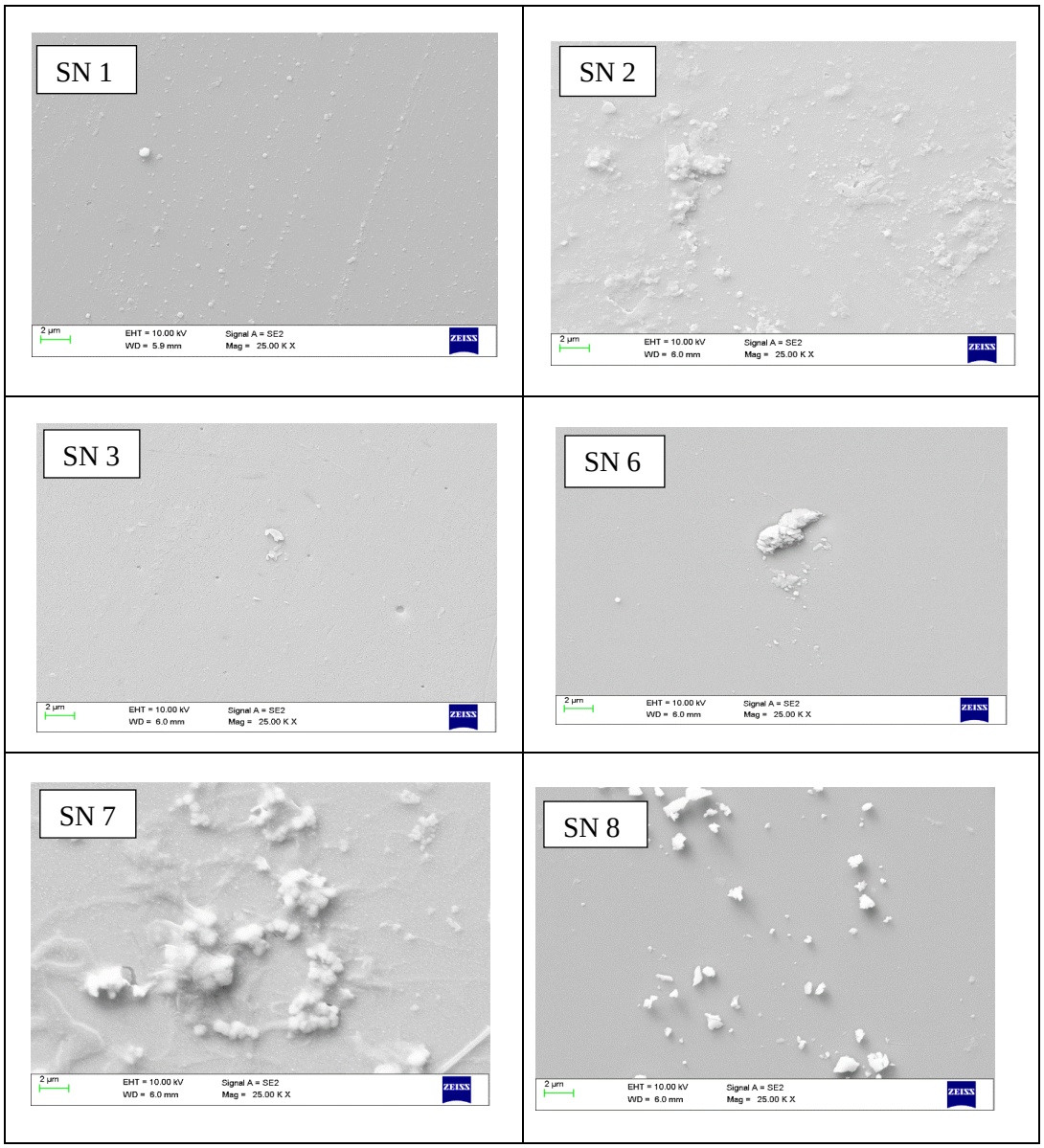
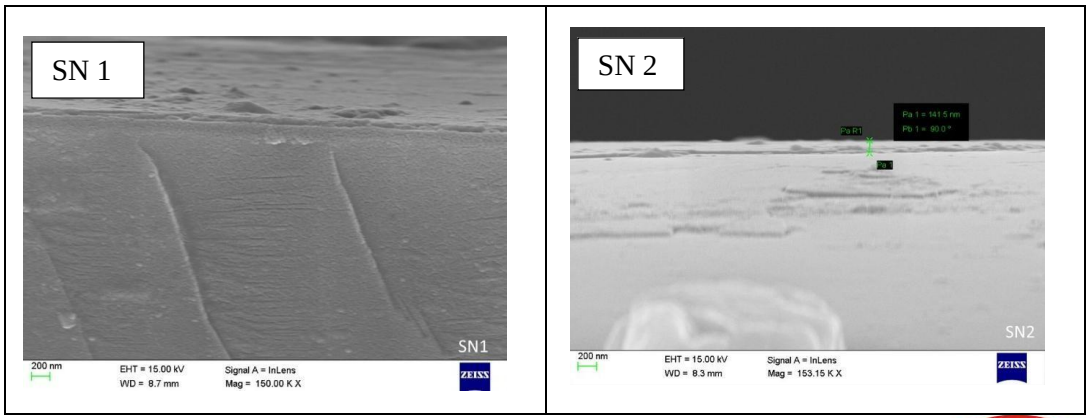
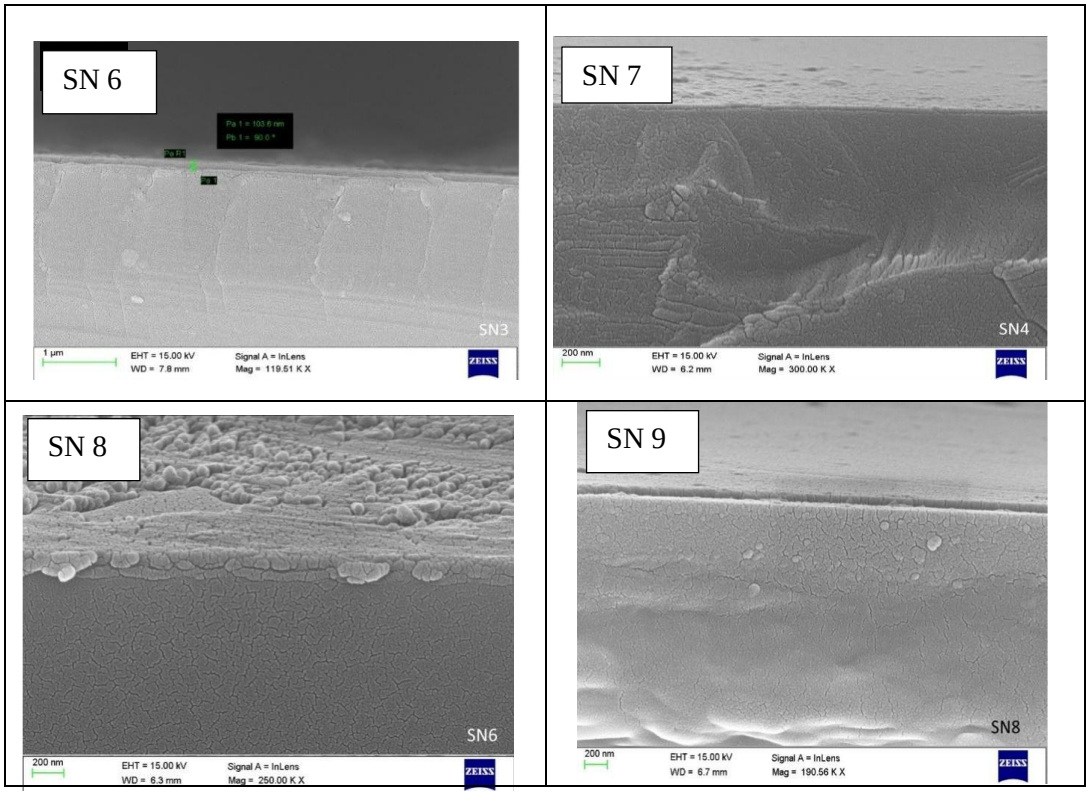


Figure 3. Cross section of the Si_3N_4 coating





Elemental composition of the Si_3N_4 (EDS)

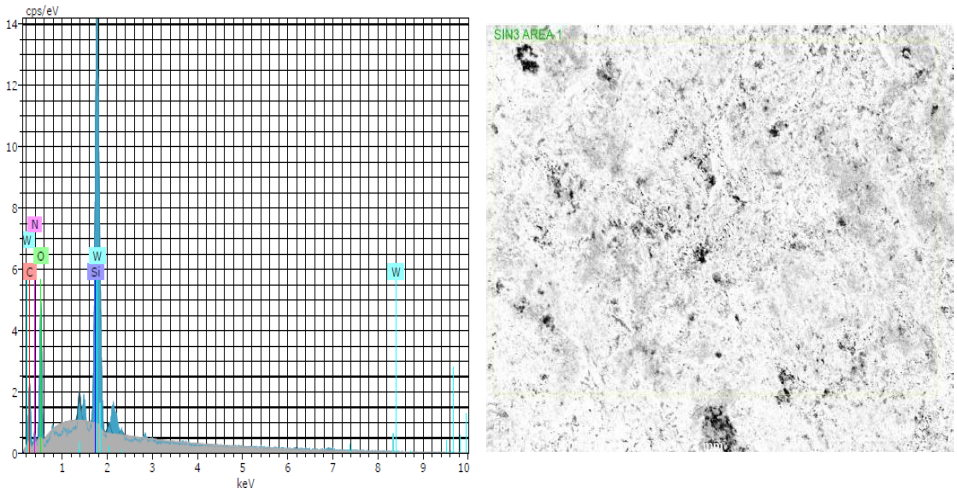


Figure.4 EDS chemical composition graph and phase contrast

Table 4 Elemental composition of the Si_3N_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

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 co efficient 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 (Si_3N_4). The AFM results shows in Table 6. Figure 6 shows the 3D AFM images of the Si_3N_4coating.

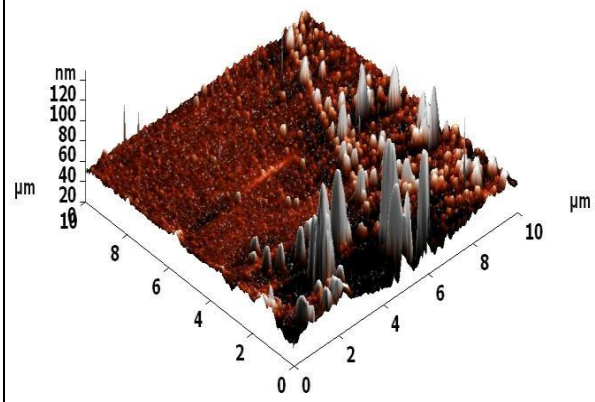
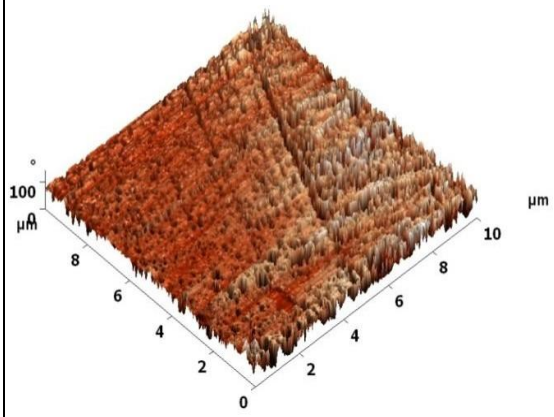
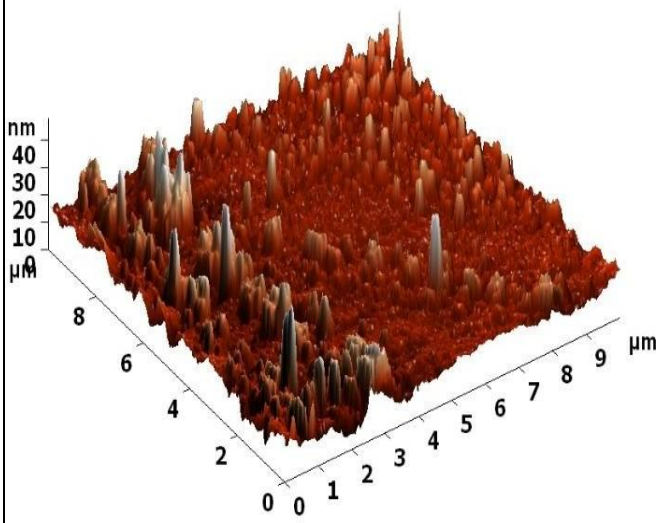
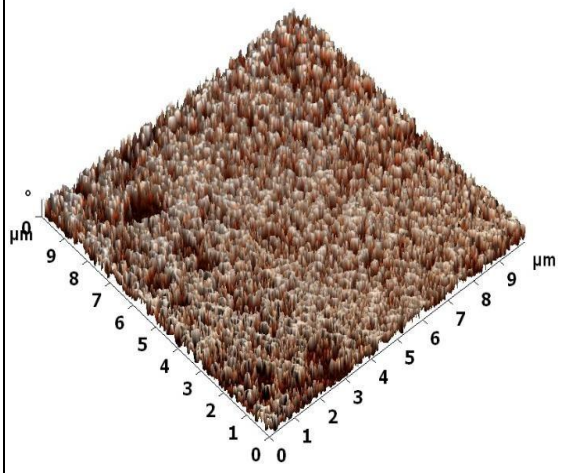
Table 5Thickness of the Si_3N_4Coatings

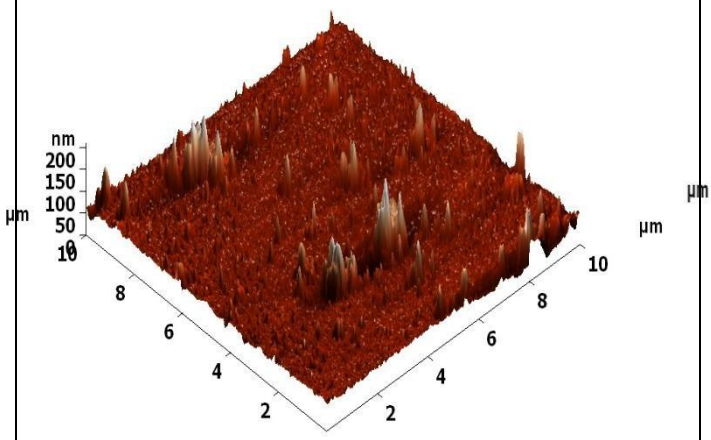
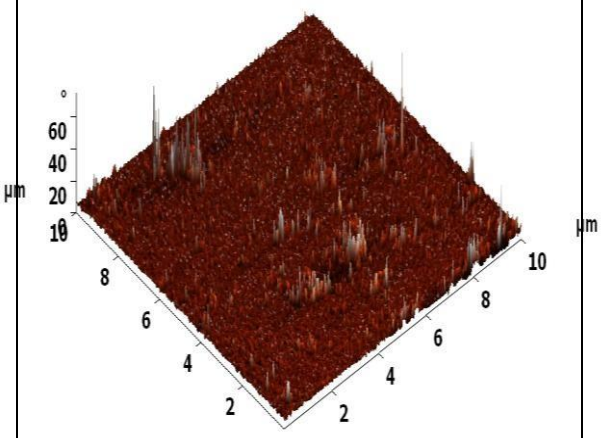
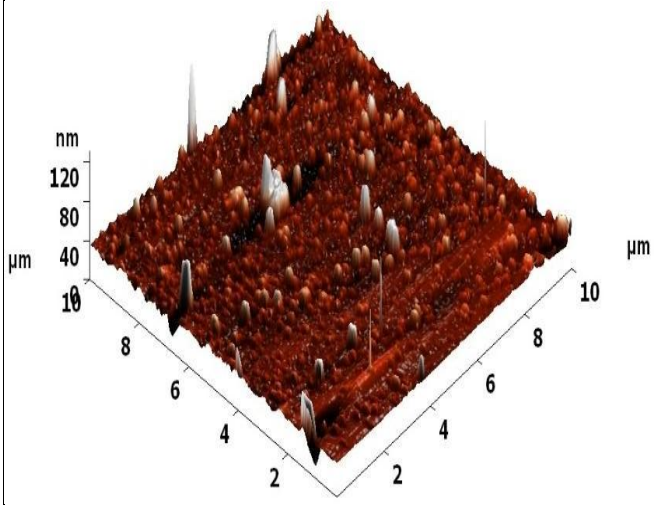
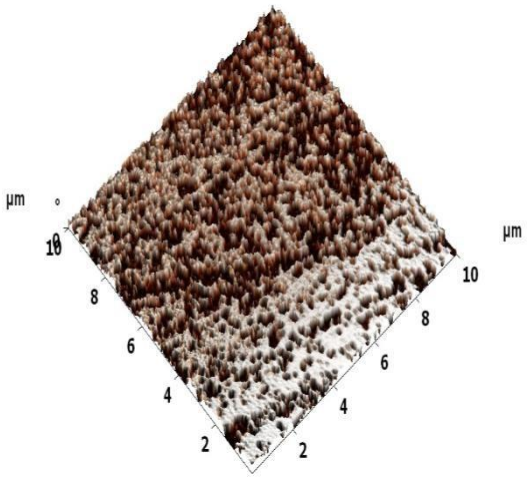
S.No	Sample ID	Thickness (nm)
1	SN1	81.2
2	SN2	98.4
3	SN3	103.6
4	SN4	45.68
5	SN6	98.46
6	SN7	62.5
7	SN8	101.2
8	SN9	75.8

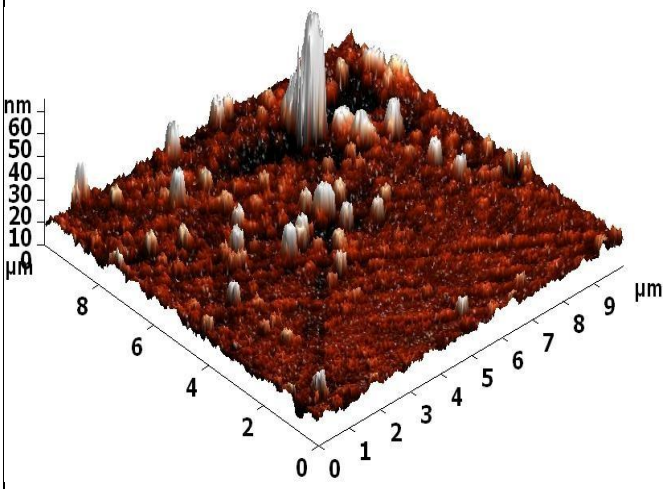
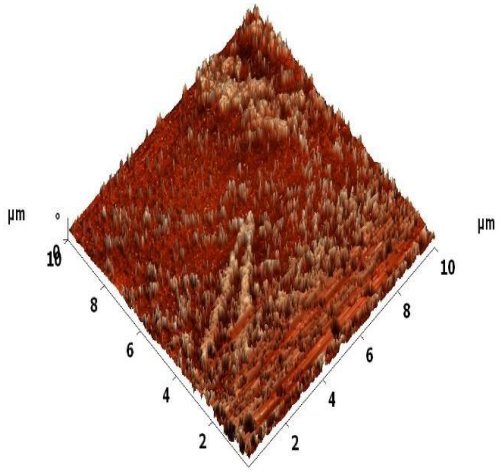
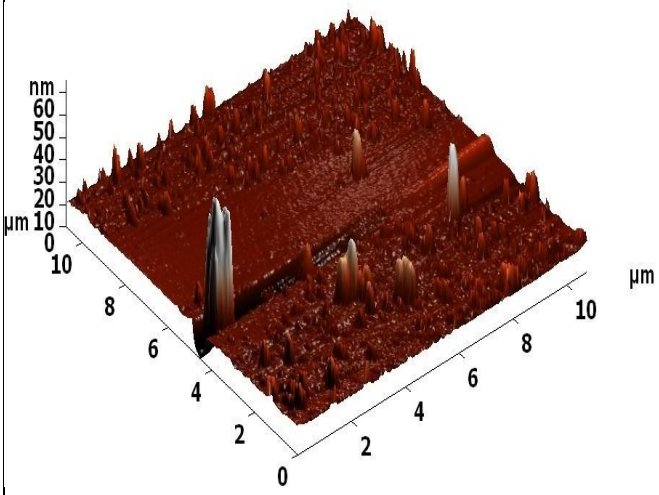
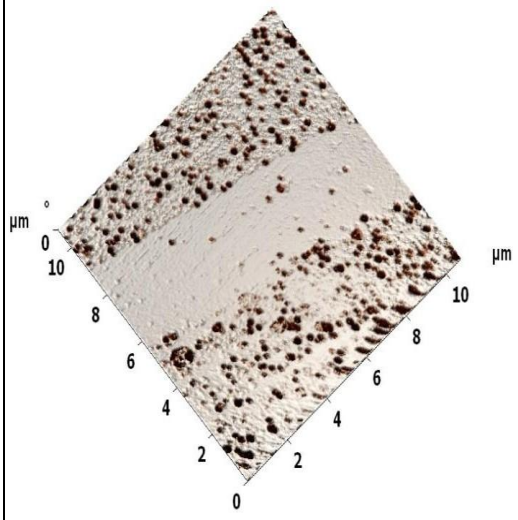
Table 6. Surface Roughness for Coated Si_3N_4 Substrate

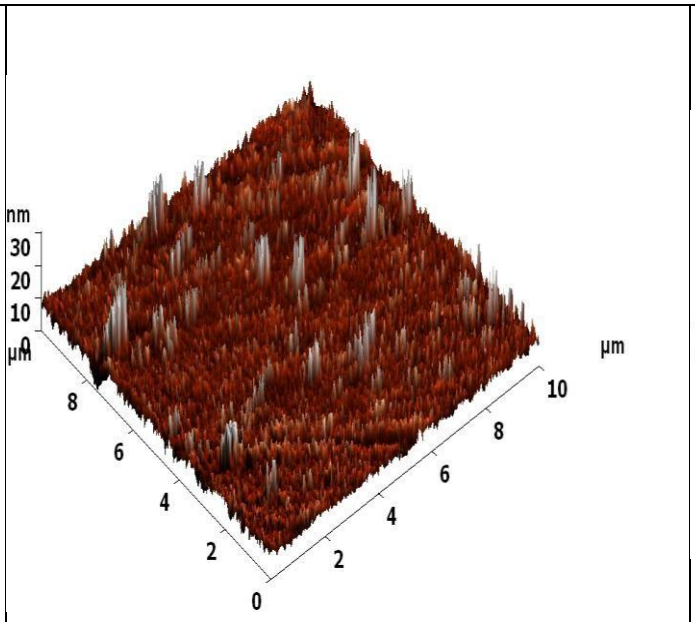
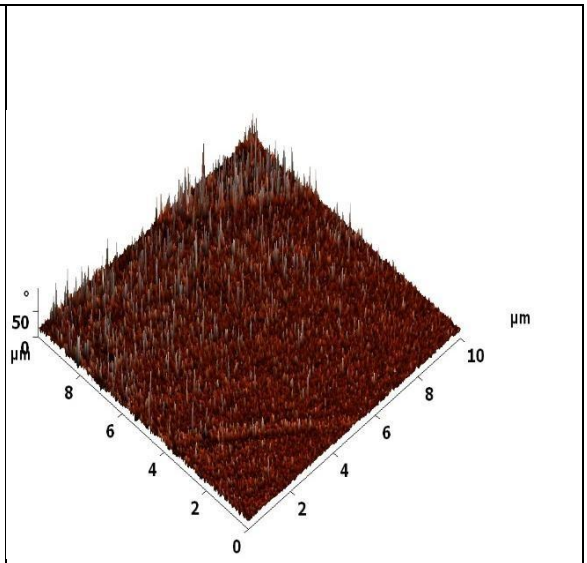
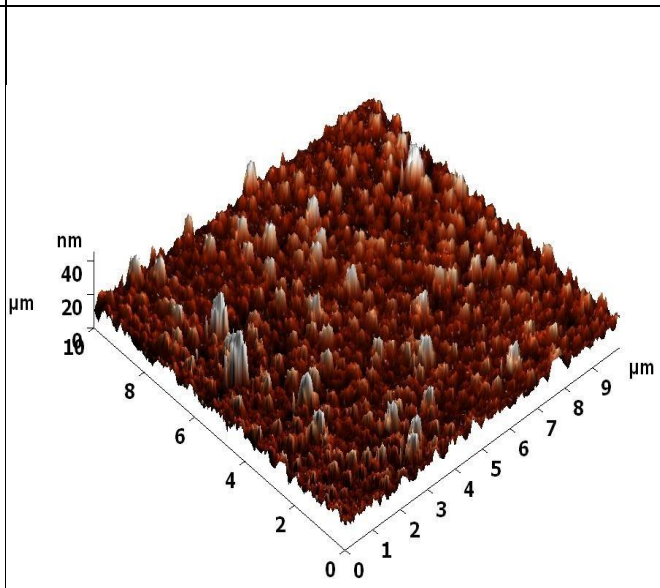
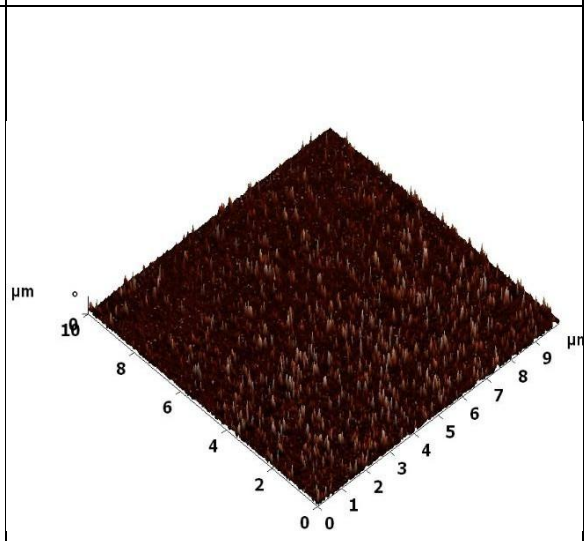
S.No	Sample ID	Surface Roughness [nm]
1	SN1	1.54692
2	SN2	2.52180
3	SN3	1.08666
4	SN4	2.94149
5	SN6	1.55059
6	SN7	1.92102
7	SN8	1.49209
8	SN9	2.90802

Figure. 5 AFM 3D Topology and Phase Contrast

Sample ID	3D-Topology	3D-Phase contrast
SN1		
SN2		

SN3	 nm 200 150 100 50 0 µm µm 2 4 6 8 10	 60 40 20 0 µm µm 2 4 6 8 10
SN4	 nm 120 80 40 0 µm µm 2 4 6 8 10	 µm µm 2 4 6 8 10

SN6		
SN7		

SN8		
SN9		

V. S/N Ratio for Thickness and SurfaceRoughness

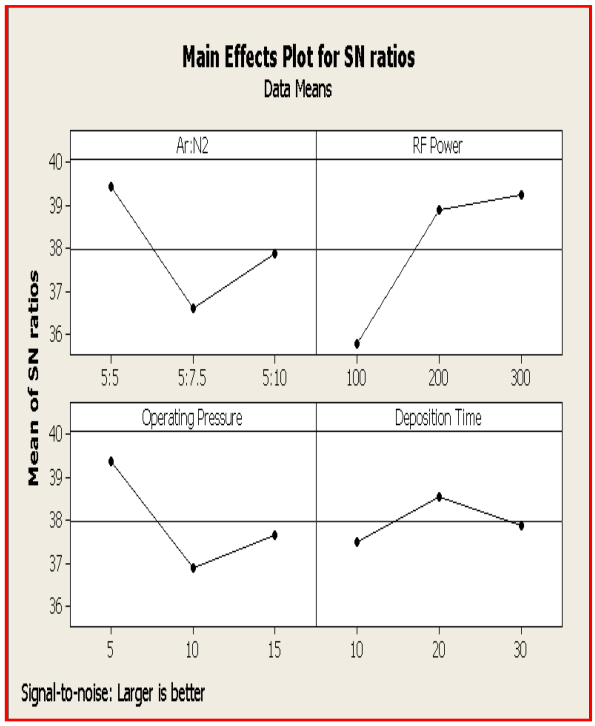


Figure.5 S/N RatioforThickness

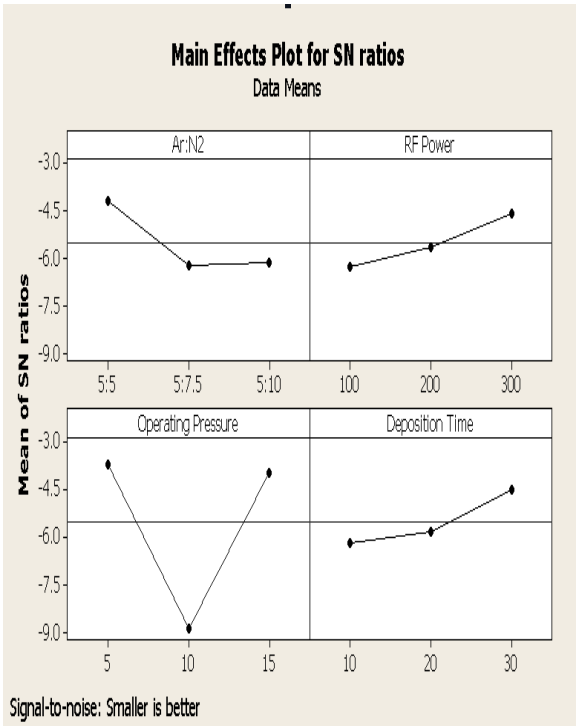


Figure.6 S/N Ratio for SurfaceRoughnes

The optimum combination parameters for producing high thickness and uniform deposition of Si_3N_4 layer were determined as deposition time 20 min, Ar:N_2 flow rate of 5:7.5 sccm, RF sputtering power of 200W, base pressure of 10mTorr.

VI. CONCLUSIONS

The silicon Nitrate (Si_3N_4) 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 characterizationresults:

1. The SN3 sample coated perfectly with smooth surface on substrate. EDS characterization also confirmed the formation of the composition ofthe Si_3N_4depositions. SN3 substrates has5.39% 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 Si_3N_4 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.

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