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A review paper on tribological and mechanical properties of ternary nitride based coatings

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Abstract

The nanotribological studies are needed to develop fundamental understanding of interfacial phenomena on a smaller scale in nanostructures used in magnetic storage systems and other industrial applications. In several fields of technical application, e.g., in the automotive or aircraft industries, the reduction of friction and wear is a very important aspect with respect to longer lifetimes, increased service intervals and lower operation costs of tools, machines and other devices or mechanically loaded elements. Many researchers have studied tribological properties of various binary thin film materials to investigate various properties like adhesion, hardness and friction coefficients according to the respective applications. The different deposition techniques they used are Unbalanced Magnetron Sputtering. Ion Beam Assisted Deposition, Physical Vapour Deposition etc. Recently ternary nitride based coatings are investigated as they exhibit improved tribological properties of three ternary nitride based coatings. This review paper is aimed to summarize tribological properties of three ternary nitride based coatings such as Titanium Aluminium Nitride, Titanium Vanadium Nitride and Titanium Molybdenum Nitride.

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1. Introduction

Thin film coatings are being increasingly used for tribological applications. In modern industrial system the major challenges are to design and produce materials having a low wear rate and low coefficient of friction for a wide range of working environments [1]. Surface engineering is a fast growing area of research because of the high industrial demands for friction control and wear resistance, coupled with enabling technology that produces new coatings with desirable tribological performance as well as mechanical properties [2]. These coatings have many tribological applications such as gas turbine bearings, diesel engine piston rings, aerospace components or forming tools, etc [3]. The elevated friction and wears generated causes essential energetic and material losses and decreases efficiency of mechanical systems.

Nitride base coatings are applied on automobile parts and used as wear resistant coatings for cutting tools [4,5]. They possess high hardness, good wear resistance, low friction and corrosion resistance [6,7,8,9,10,11]. Development of complex ternary nitride coatings has attracted significant research and industrial interest in the last few years, to get better film property like high hardness, good wear resistance and excellent corrosion protection, high melting point etc. Ternary nitride based coatings are achieved by addition of a third material such as Vanadium (V), Aluminium (Al), Molybdenum (Mo), Silicon (Si), Zirconium (Zr) etc. to a binary nitride compound (e.g. TiN, MoN, VN, ZrN, CrN, SiN, etc.). A small quantity of the third material changes the morphology, structure and bonding of the film coating [12,13]. The objective of this paper is to review tribological properties of three ternary nitride based coatings namely Titanium Aluminium Nitride (TiAlN), Titanium Vanadium Nitride (TiVN), and Titanium Molybdenum Nitride (TiMoN).

2. Titanium Aluminium Nitride (TiAlN) coatings

Titanium Nitride (TiN) is hard protective coating, its oxidation resistance at elevated temperatures sets a critical limitation to the operation conditions. Especially during the operation of cutting tools coated with TiN, the temperature increase induced by the friction between the tool and its counter surface at the contact is inevitable in most cases. TiN has a thermodynamically favoured tendency to replace the bonded nitrogen with gas oxygen molecule, followed by the out-gassing of nitrogen. This oxidation process is activated thermally between 500°C to 700°C, leaving oxidized titanium with reduced hardness on the substrate. Along with its bad wear resistance and adhesion, the titanium oxide layer is either removed or delaminated soon, exposing fresh TiN coating (to be oxidized repeatedly) or substrate leading to tool failure [14]. In order to improve the oxidation behaviour of TiN, in the mid 1980's Aluminium (Al) has been added forming the Titanium Aluminium Nitride (TiAlN) ternary system [15].

The Al has higher mobility than Ti moving through the parent phase and the oxides of titanium and aluminium, which makes Al to be oxidized more in amount than Ti. The oxidized Al forms a dense aluminium oxide layer on the surface of the coating, where the layer can effectively retard further penetration of oxygen and extend the lifetime as compared to pure TiN [14,16]. Titanium-Aluminium-Nitride is a well-studied ternary system from the 1980's. The first reported phase was the Ti_2AIN in 1963, which is characterized by the high temperature abrasion resistance and good thermal-electrical conductivity [17].

Additional advantages besides the enhanced oxidation resistance by adding Al into TiN were also reported in the late 1980's. One was the improvement of mechanical properties of the coatings with respect to the binary counterparts TiN and AlN. Many groups observed increase of the hardness of TiAlN coatings during heat treatment above 700° C [18] and around 1990s, mechanisms were proposed to explain this behaviour. The mechanisms were based on the age hardening effect triggered by a thermodynamic phenomenon, known as the spinodal decomposition. Spinodal decomposition creates domains with different composition in the coatings retarding the motion of dislocations [19].

In several industrial applications such as milling and forming processes coatings are required to have high hardness and elasticity as well as high oxidation resistance, which enables such coatings to be employed even under higher temperatures [20]. Yeung W. Y. *et al.* [21] fabricated ternary nitride coatings (TiAlN) through reactive magnetron co-sputtering at nitrogen deposition pressures of 0.053 and 0.128 Pa. The effect of nitrogen pressure on the hardness was studied and hardness of 18.6 GPa at 0.053 Pa and 13.6 GPa at 0.128 Pa was observed.

TiAlN coated cutting tools have shown excellent wear resistance in machining sticky metals such as aluminium alloys and austenite stainless steel [22]. AlTiN has exceptionally high chemical resistance; the superiority of the AlTiN coating against aluminum is confirmed by wear rates estimations by Yucel B. *et al.* [23]. Chu K. *et al.* [24] prepared SS (Ti,Al)N films on Si wafers and AISI M42 tool steels (for tribological measurements). They have studied mechanical and tribological properties of SS (Ti,Al)N films as a function of negative bias voltage prepared at room temperature by reactive close-field unbalanced magnetron sputtering in an argon and nitrogen gas mixture. It was found that the nanohardness and residual stress were clearly related to the negative bias voltages.

Ananthakumar R. *et al.* [25] prepared TiN/TiAlN multilayered coatings with bilayers lengths of 20–30nm and thickness of 2μ m by DC reactive magnetron sputtering using 99.9% pure titanium and titanium aluminum alloy target. Substrates were cleaned in ultrasonic bath using acetone and trichloroethylene. The frictional behaviour of the coatings under a normal load of 3.924N at room temperature is shown in figure 1(a). Sliding friction for TiN/TiAlN coated films shows a relatively lower value of about 0.25 compared to the 0.40 and 0.45 for the single layer coatings and bare substrate. TiN/TiAlN multilayered coatings show the lowest coefficient of friction compared to single layer and bare substrate. In figure 1(b) a large difference can be seen in wear rate and found to be the lowest for the TiN/TiAlN multilayer coatings.



Figure 1: (a) Friction coefficient of the TiN/TiAIN multilayer coatings. (b) Wear rate for TiN/TiAIN multilayer coatings [25].

Shum P.W. *et al.* [26] deposited Ti_{1-x}Al_xN films on n-type Si(1 0 0) wafers and M42 tool steels at room temperature by reactive close-field unbalanced magnetron sputtering from high purity of three titanium (Ti) and one aluminium (Al) targets. The base pressure in the vacuum chamber was 3×10^{-4} Pa, and the working pressure, consisting of Ar and N₂ with a constant flow was set at 0.27Pa during all depositions. They studied the variations of steady-state friction coefficient of Ti_{1-x}Al_xN films as a function of x as shown in figure 2(a). At a normal load of 2N, the friction coefficient of Ti film (x =0.09) led to a sudden increase of friction coefficient to 0.70. With further increase in the Al content of the film, the friction coefficient maintained relatively lower values of 0.55–0.65. Figure 2(b) represents the specific wear rate of Ti_{1-x}Al_xN films as a function of the Al content. Overall, a higher friction coefficient resulted in a higher wear rate. Table 1 summarizes the investigation done by various research groups to study effect of different parameters as per deposition techniques on tribological and mechanical properties for TiAlN system.



Figure 2: (a) Steady state friction coefficient and (b) specific wear rate of the $Ti_{1,x}Al_xN$ films on M42 steel substrates as a function of Al content (x) at applied loads of 2, 5 and 20N [26].

Research Group/s	Deposition Technique	Variable Parameters	Properties Examined	
(Year)			Tribological	Mechanical
Qi Z.B. <i>et</i> <i>al</i> .(2013)[27]	Cathodic arc ion plating	Tribo Test Temperature: 25°C to	COF:0.78 to 0.70	Hardness: 22.45 GPa to 12.38 GPa
Radhika R. <i>et al.</i> (2013)[28]	Physical Vapour Deposition (PVD)	Counter bodies: 100Cr6 steel, SiC and Al ₂ O ₃	COF: 0.3 to 0.45 Wear:4×10 ⁻¹⁰ ,1.7×10 ⁻⁷ , 2.3×10 ⁻⁷	NIL
Ipaz. L. <i>et al.</i> (2013)[29]	Pulsed dc magnetron co-sputtering	90:10 and 50:50 (Ar:N ₂ ratio)	COF: 2.8 to 3.5	NIL
Yucel B. <i>et al.</i> (2013)[30]	Cathodic Arc Physical Vapor Deposition (CAPVD)	P _{N2} : 1 Pa, Bias voltage: -100 V.	Wear: 0.004 $\mu m^3 N^{-1} m^{-1}$	Hardness: 3291±118 HV
Xin W. <i>et</i> <i>al</i> .(2013)[8]	Cathodic-arc evaporation	T: > 450°C Counter bodies: WC/6%Co, Alumina, steel l, SiC, Si ₃ N ₄	COF:0.52,0.87,0.7,0.77,0.85	NIL
Zhao Y.H. <i>et al.</i> (2012)[31]	Pulse biased arc ion plating	Film Thickness:1.08 to 2.53µm	NIL	Hardness: 27.8 to28.1(GPa) Elastic modulus: 344.7 to 337.3(GPa)
Ricardo D. T. <i>et al.</i> (2010)[32]	PVD cathodic arc evaporation (CAE)	Film Thickness: 3 to 3.3 µm	Wear:2.40×10 ⁻⁵ , 1.24×10 ⁻⁵ , 1.05×10 ⁻⁵	Hardness: 9.5 and 10.5GPa
Zhou Z. <i>et al.</i> (2010)[33]	combined steered cathodic arc/unbalanced magnetron (UBM) sputtering	Tribo Test Temperature: 25°C, 300°C to 635°C	COF:0.46 to 1.03 Wear:-90×10 ⁻¹⁵ to 2.3×10^{-15} m ³ N ⁻¹ m ⁻¹	NIL
Mo J.L. <i>et al.</i> (2007)[34]	Multiple arc vapour deposition technique	DC-bias voltage: -50 V to-150 V.	COF: 0.70 ot 0.85	Hardness: 35.72±9.84 GPa Elastic modulus: 460.35±80.33 GPa
W. Grzesik. <i>et al.</i> (2006)[35]	PVD	Load: 10 to 30N Speed: 0.5 to 1.5 m/s	COF: 1.01to 0.72, 1.12 to 0.73 and 1.30 to 0.82 Wear: 4.2 to 5.2×10 ⁻⁶ mm ³ /(mN)	NIL
Ichimiya N. <i>et al.</i> (2005)[36]	cathodic arc ion plating	P_{N2} : 2 to 5 Pa	COF: 0.91	NIL
Wuhrer R. <i>et</i> <i>al</i> .(2004)[37]	Reactive magnetron co- sputtering	P_{Ar} & P_{N2} : 0.32Pa and 0.053 Pa	NIL	Hardness: 2400 HV
Ohnuma H. <i>et</i> <i>al</i> .(2004)[38]	Cathodic arc ion plating method	Atomic ratios (Ti:Al) Test T: 300, 473,673,873 K	COF is lower values	Nano-hardness decreased with increasing Al
PalDey S. <i>et al.</i> (2003)[39]	A cathodic arc vapor deposition system	N ₂ : 0.66, 1.33, 1.99, 2.66 Pa	COF: 0.091,0.075,0.093 and 0.087	Hardness: 2783±25 HV
Daniel K. <i>et al.</i> (2012)[40]	PVD techniques of ARC and SARC	N ₂ flow rate:120cm ³ /m	COF: TiN=0.82and TiAlN=0.42	Hardness: TiN=31.33 GPa TiAlN=26.7 to 29.2 GPa
Hiroyuki H. <i>et al.</i> (2000)[41]	Arc ion plating method	P _{N2} : 3.3 Pa	NIL	Hardness: TiN=200HV TiAIN= 3100HV
Vancoille E. <i>et al.</i> (1993)[42]	Steered arc ion plating	Tribo Test Temperature: 300°C to 900°C	NIL	Young's modulus(GPa): 201±50 to 298±72 Hardness: 733±268 to 2419±386HVN

Table1: Review of Titanium Aluminum Nitride thin films

3. Titanium Vanadium Nitride (TiVN) cotings

Titanium vanadium nitride is technology important thin film coatings used in a diverse range of areas such as the packaging industry, transparent barrier coatings and microelectronics. The level of intrinsic stress in the film controls the films adhesion to the substrate and the microstructure controls the usefulness of the film [43]. Information in the literature concerning titanium vanadium nitride has shown the superior performance of such coated ternary compounds to those of both pure TiN and VN, with respect to their hardness and tribological environments [44].

The materials richest in titanium and vanadium were produced through a combustion process time in 3h and a diffusion process time in 9h respectively. Reactive milling is a good method to obtain these types of compounds because it is inexpensive if compared with deposition and thermal techniques. The employed characterization methods indicate that the materials exhibited nanometer particle sizes, high sinter abilities and high micro hardness [44].

Knotek O. *et al.* [45] found that Ti-V-N magnetron-sputtered thin films have high hardness with excellent thermal stability. The coating composition is modified simply by altering the target composition and the process parameters (partial nitrogen pressure, substrate temperature and substrate bias). The wear test results show the best wear resistance for the coatings at 29 %V [46].

Table 2 summarizes the investigation done by various research groups to study effect of different parameters as per deposition techniques on tribological and mechanical properties for TiVN system.

Research Group/s	Deposition Technique	Variable Parameters	Properties Examined	
(Teal)			Tribological	Mechanical
Teerawit D. <i>et al.</i> (2013)[47]	Reactive DC magnetron co-sputtering	Vanadium(V) sputtering current I_V (A): 0.4,0.6,0.8 and 1	NIL	structure, surface and cross-sectional morphologies
Ouyang J.H. <i>et al.</i> (2007)[48]	cathodic arc ion-plated	Tribo Test Temperature: 200 °C to 300°C, 400 °C,500°C, 600 °C to 700°C	COF: 1.01, 1.08±0.06, 1.06±0.05, 0.68±0.02, 0.55±0.02 and 0.61±0.04 Wear:- 1.69×10 ⁻⁷ to1.31×10 ⁻⁵ mm ³ /Nm	NIL
Ichimiya N. <i>et al.</i> (2005)[36]	cathodic arc ion plating	P _{N2} : 2 to 5 Pa	COF:- 0.5 to 0.6	Hardness: 2600 HV
Yeung W. Y. <i>et al.</i> (2006)[21]	Reactive magnetron co- sputtering	$P_{\rm N2}{:}~0.053$ and 0.128 Pa	NIL	Hardness: 13.6 to18.6 GPa Elastic Modulus: 196 to 264 GPa
Luo Q. et al. (2006)[49]	Unbalanced magnetron reactive sputtering	P _{Ar} :P _{N2} =50:30	COF: 0.4 Wear: $2.3 \times 10^{-17} \text{m}^3 \text{N}^{-1} \text{m}^{-1}$	Hardness: 28.3 GPa
Ouyang J.H. <i>et al.</i> (2004)[50]	Cathodic arc ion plating (CAIP)	Load(N): 20 to 70	COF: 0.68 to1.08 Waer: 4.17×10^{-8} to 2.25 × 10^{-6} mm ³ /Nm	NIL
Hiroyuki H. <i>et al.</i> (2000)[40]	Arc ion plating method	P _{N2} : 3.3 Pa	NIL	Hardness: TiN=2000 TiVN=2400 HV

Table2: Review of Titanium Vanadium Nitride thin films

Ti-Mo-N is also an effective alternative ternary film coating material to improve the mechanical properties of TiN. Ti-Mo-N film had a lower friction coefficient and wear rates than TiN films. The hardness is significantly increased with the introduction of nitrogen gas, and reaches a maximum value of approximately 30 GPa at $f_{N2}=0.3$ ccm. The hardness is slightly reduced by further increase in N₂ flow rate [51,52].

Wiemer C. *et al.* [53] worked on samples of TiMoN in a wide range of chemical compositions deposited by reactive magnetron sputtering from a dual cathode. The variations of the chemical composition caused by the changes in the power of the targets and nitrogen partial pressure are related to the different enthalpies of formation of the binary nitrides. In $Ti_{1-x}Mo_xN_y$ system, the formation of a single face centered cubic phase for molybdenum (Mo) up to 0.79 is found to be possible by r.f. reactive magnetron sputtering. The presence of titanium in the metallic sublattice, and the similarity between the lattice parameter of cubic TiN and MoN allows the synthesis of the well-ordered structure. The new TiMoN system presents micro hardness values 40% superior to TiN. Regent F. *et. al.* [54] found that the Ti-Mo-N film hardness (H) varies with nitrogen pressure (P_{N2}), the values observed are at P_{N2} =0.15 Pa, H= 40 GPa and P_{N2} =0.25 Pa, H=38 GPa.

Yang Q. *et al.* [55] fabricated TiMoN coatings on Ti6Al4V substrates by reactive magnetron sputtering using two pure Ti and two pure Mo targets in a closed-field unbalanced magnetron system with the Ar flow rate controlled by a mass flow controller and the N₂ supply regulated by an optical emission monitoring system. They found that the coefficient of friction is a function of molybdenum concentration (X_{Mo}) as show in figure 3(a). It is evident that the increase in molybdenum concentration effectively lowers the coefficient of friction of the coatings when tested against a WC-Co counterpart. The coefficient of friction first decreases dramatically as X_{Mo} increases, and then becomes relatively stable when X_{Mo} is in the range of 0.3–0.6. In this range, the coefficient values (0.4–0.5) are less than half of that for TiN coating (1.03). The wear rates of the TiMoN coating as a function of X_{Mo} is shown in figure 3(b). Figure 3(b) shows that Mo alloying significantly improves the coating wear resistance. The wear rates decrease monotonically with X_{Mo} up to 0.5, followed by a slight increase at X_{Mo} = 0.57. The wear rates in figure 3(b) were derived from 1000 m sliding tests for coatings with X_{Mo} less than 0.4; when X_{Mo} is larger than 0.4 because of the excellent wear resistance of the coatings, 2000 m sliding tests were performed to alleviate the difficulty in wear rate measurement. For TiMoN coatings with X_{Mo} in the range of 0.4–0.6, their wear rates are in the order of 10⁻⁷ to 10⁻⁸ mm³/(Nm), only 2.5–4% of that for the TiN coating.



Figure 3: (a) & (b) Mo alloying effectively reduces the coefficient of friction and wear of the TiMoN coatings, which were measured by pin-ondisc dry sliding tests with a WC-Co ball as the counterpart [55].

Conclusion: Nitride based ternary thin films like TiAlN, TiVN and TiMoN has great demand in industrial applications, as it gives better film properties like high hardness, low coefficient of friction, good wear resistance and excellent corrosion protection, high melting point etc. as compared to binary nitride compounds (e.g. TiN,

MoN, VN). The frictional behavior of TiN/TiAlN coatings under a normal load of 3.924N at room temperature shows a relatively lower value of about 0.25 compared to the 0.40 and 0.45 for the single layer coatings and bare substrate. TiN/TiAlN multilayered coatings show the lowest coefficient of friction compared to single layer and bare substrate [25]. Adding small amounts of Al into the growing TiN film (x =0.09) led to a sudden increase of friction coefficient to 0.70. With further increasing the Al content in the film, the friction coefficient maintained relatively lower values of 0.55–0.65 [26]. The TiVN coating exhibits a friction coefficient of 0.68–1.08 at room temperature, depending on load and frequency. The wear rate of TiVN coating and alumina ball are located in the range of 4.17×10^{-8} to 2.25×10^{-6} mm³/Nm, depending on load and frequency [49]. There is a lot of scope to study the tribological properties of TiMoN film coating for various industrial applications, such as cutting tools, milling and forming processes, molds. When tested on a pin-on-disc test against Counter bodies WC–Co pin, the friction coefficients of TiMoN coatings were found to decrease with the increase in Mo atomic fraction, with the lowest friction coefficient values of 0.4 to 0.5 being only one-half that of TiN coating (1.03) [55].

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