Effect of N_2 Partial Pressure on The Structural and Mechanical Properties of Tan Films

Mehdi Manouchehrian*

Department of Physics, South Tehran Branch, Islamic Azad University, Tehran, Iran

ARTICLE INFO

Article history:

Received 31 May 2016
Accepted 28 September 2016
Available online 25 December 2016

Keywords:

Tantalum Intermetallic
Hardness
Partial pressure
Friction coefficient
Wear

ABSTRACT

TaN films with different N_2 partial pressures were deposited on 304 stainless steel using the magnetron sputtering method. The effect of gas pressure on the mechanical properties, morphology, and phase structure of the films is investigated by X-ray diffraction (XRD) analysis, atomic force microscopy (AFM), nanohardness testing, friction coefficient measurements, and wear mechanism study. The XRD results confirmed that increasing N_2 partial pressure does not affect the formation of the new phases but the intensity of the peaks increased. AFM images showed that surface roughness and grain size increased with increasing N_2 partial pressure. It was found that hardness decreased as N_2 partial pressure increased. From the friction coefficient measurements, it could be inferred that the friction coefficient increased as N_2 partial pressure increased. The images of the samples after wear test showed that wear resistance decreased with increasing N_2 partial pressure. The results show that the partial pressure of N_2 has a great influence on the mechanical properties

1. Introduction

Transition metal nitride films, such as CrN, TiN, TaN, etc., have been utilized for cutting and drilling tools due to their excellent hardness, wear, and corrosion resistance [1-4]. TaN films are used in a wide variety of applications such as thin film resistors, diffusion barriers in microelectronic [5-9], and surface protective coatings [10,11]. There are valuable investigations on the mechanical properties and thermal stability of TaN films, indicating its potential application as hard coatings [10,11]. However, the overall published reports for TaN are for less than other nitrides such as TiN and CrN. In the physical vapor deposition, PVD, TaN films show a variety of compound solutions, including BCC α-TaN, hexagonal γ TaN, Ta_2N, WC structure, cubic NaCl TaN, tetragonal Ta_4N_5 and orthorombic Ta_3N_5 [12-13]. The structures of TaN films prepared by PVD method depend intimately on the deposition technique and the process parameters. Multilayer structure and related mechanical and corrosion behaviors were also intensively studied [14]. In this work, we propose to study the effect of N_2 partial pressure in TaN films in terms of the microstructure, surface morphology and mechanical properties of the films.

2. Materials and methods

The tantalum nitride films with 800 nm thickness measured by Quartz digital thickness gauge, were deposited on 304 stainless steel

* Corresponding author:
E-mail: mehdi_manoochehrian@yahoo.com

substrates by magnetron sputtering technique. The target diameter and thickness were 50 mm and 6 mm, respectively, and the distance of target from substrate is 100 mm. The substrates were cut into 10mm ×10mm slabs and then cleaned with acetone and ethanol in an ultrasonic bath for 30 min. The vacuum chamber base pressure was 2×10-5 mbar. Argon (99.999%) and nitrogen (99.999%) were introduced into the chamber through mass flow controllers, which were used as the sputtering and reactive gases, respectively. The voltage and substrates temperature were fixed at 220 V and 300 K respectively during the deposition. The TaN films were deposited on 304 stainless steel with different N_2 partial pressures which isas summarised in table Table 1.

Table 1. Flow rate of N2 and Ar used for depositing TaN films

Samples	Ar(%)	N_2(%)
C_1	90	10
C_2	80	20
C_3	70	30

The phase and crystalline structure of the films were characterized by X-ray diffraction (XRD) analysis using a Philips-pw 1800 with Cu(-Kα) radiation (λ=0.15406) which was operated at 40 kV and 30mA. The scanning angle was from 2θ=20° to 2θ=70° with a step size of 0.02° and a measuring time of 1.25 s per step. The topography and roughness of the films were investigated using an atomic force microscope (DI, Dimension 3100) in contact mode. Hardness and Young's modulus were measured using a load-controlled nano-indentation system (UNI 2000) with a Berkovich indenter. The load dwelling time was 30 s while the indentation depth was 200 nm. The measurements on Eeach sample was were measured carried out for five times, and the average values as well as the deviation were calculated. Friction coefficient and wear tests were performed using a pin-on-disk tribometer. A Si3N4 ball with a diameter of 6 cm was used as the counterpart. The Normal normal loading was 10 N and the relative sliding speed was 0.08 m/s. The tests were carried out under atmospheric pressure at 25 °C and 50% relative humidity. Prior to the experiments, the samples were ultrasonically cleaned for 10 min in ethanol, in order to remove substances that might affect the results. Images of the wear track mechanism in the samples with different N2 partial pressures were examined by scanning electron microscope.y (SEM, Hitachi S-4160, Japan).

3. Results and discussion

Figure 1 shows the XRD patterns of the samples with different N_2 partial pressures. In all XRD patterns, in addition to the peaks denoted with star which are related to 304 stainless steel substrate, the peaks of TaN(111), TaN(200) and TaN(220) with FCC structure are observed at 2θ=35.5°, 2θ=41.6° and 2θ=60.4°, respectively. It can be observed in Fig. 1 that TaN(111), TaN(200), and TaN(220) peaks intensity in fig 1 increased with the increase of N_2 partial pressure which indicates The effect of N2 partial pressure on the microstructure of different films is shown in figure 2. The surface topography of the films is performed using height AFM for a scanned area of 2μm×2μm. As can be observed, the AFM images of the samples shows a columnar structure in which the round grains with the nearly equal sizes are distribution distributed with distance to from one another on the an improvement in the crystalline structure of the films. When the partial pressure of N2 further increased (sample C3), TaN(200) and TaN(220) peaks intensity increased, while the TaN(111) peak intensity decreased, which indicates that the preferred orientation of the TaN films have has changed. surface. Table 2 shows the values of average (Ra) and the root mean square roughness (Rms) as well as grain size for of the samples. The grain size in the AFM images increases as a function of the N2 partial pressure. Also, the surface roughness of the films increased with the N2 partial pressure because of the increase of the grain size as shown in 2D images of figure2. Figure 3 shows the hardness and Young's modulus of the films as a function of N_2 partial pressure. The hardness and Young's modulus decreased with the increase of N_2 partial pressure. Overall, factors like surface roughness, N_2 partial pressure and grain size have the greatest impact on hardness and

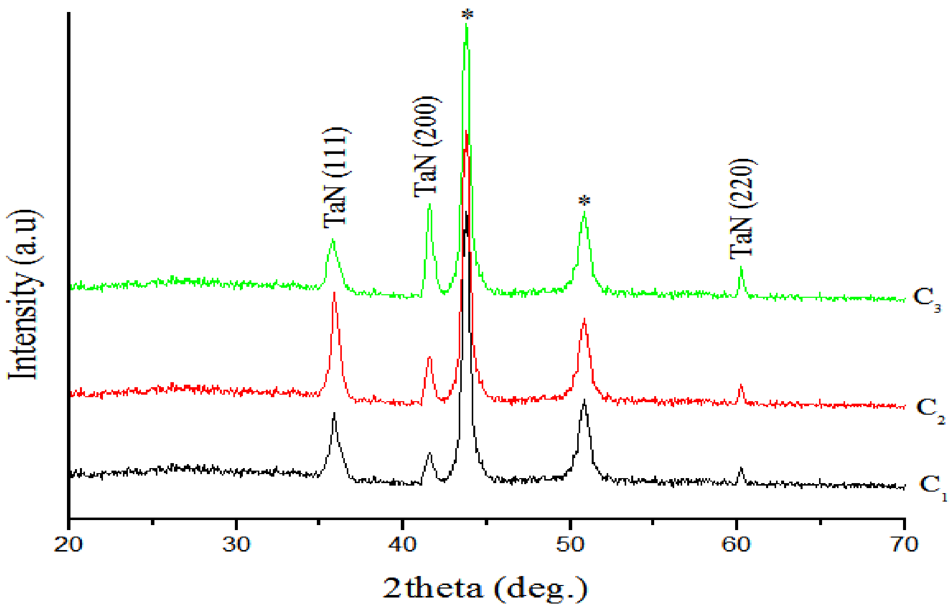

Fig. 1. XRD patterns of the TaN films prepared at different partial pressures of N_2

Table 2. Variation of the average roughness and root mean square roughness as well as grain size

Samples	surface roughness (Å)		Grain diameter (nm)
	Rms	Ra	
C_1	8.3	6.1	138
C_2	10.1	7.9	146
C_3	16.1	13.2	180

Young's modulus. According to the hardness values obtained in this study, it can be said that grain size has the strongest effect on hardness. Decreasing the grain size according to the Hall-Petch equation causes the increase in hardness and Young's modulus [15].

The plastic deformation resistance (H^3/E^2) is a strong indicator of a coating resistance to plastic deformation in the elastic/plastic plate contacts [16]. The high H^3/E^2 of 0.175 was obtained for sample C_1. Hence, it is expected that sample C_1 shows better wear resistance. The elastic recovery was calculated by the following equation:

$$R= h_{max}-h_r/h_{max} \quad (1)$$

Where h_{max} and h_r are the maximum and residual displacements, respectively. Elastic recovery (R) [17] in TaN films was evaluated by load-penetration depth curves of indentations for each film, as show in fig 4. The values decreased from 37% to 28% with increasing N_2 partial pressure. Figure 5 shows the patterns of the friction coefficient versus distant for all samples. It can be seen that the friction coefficient increased with increasing N_2 partial pressure. Generally, the friction coefficient depends on two factors, namely surface roughness and hardness. Sample C_1 had the lowest friction coefficient because of its less surface roughness and more hardness compared with other samples.

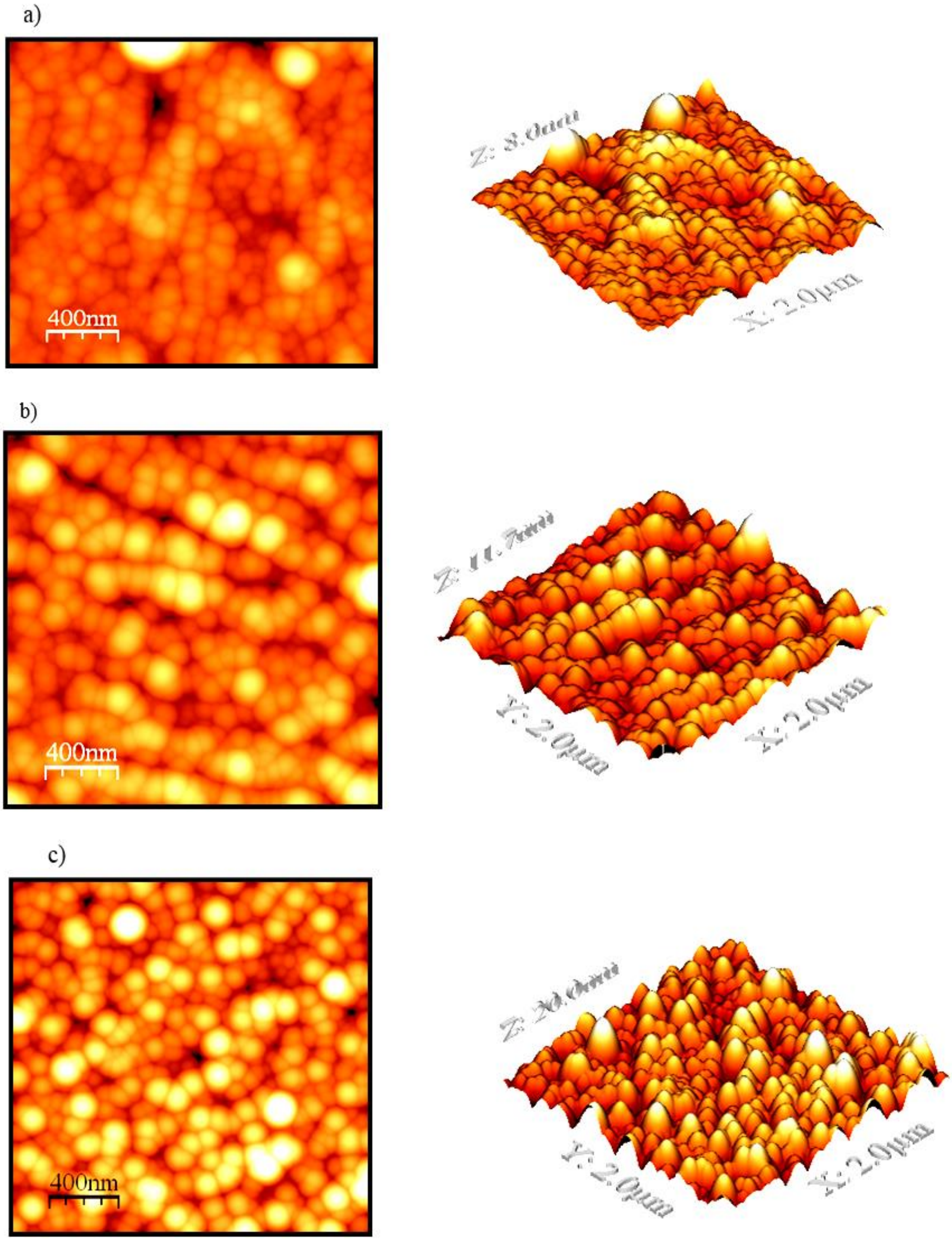

Fig. 2. AFM images of the TaN films prepared at different partial pressures of N_2:(a) 10%, (b) 20% , (c) 30%.

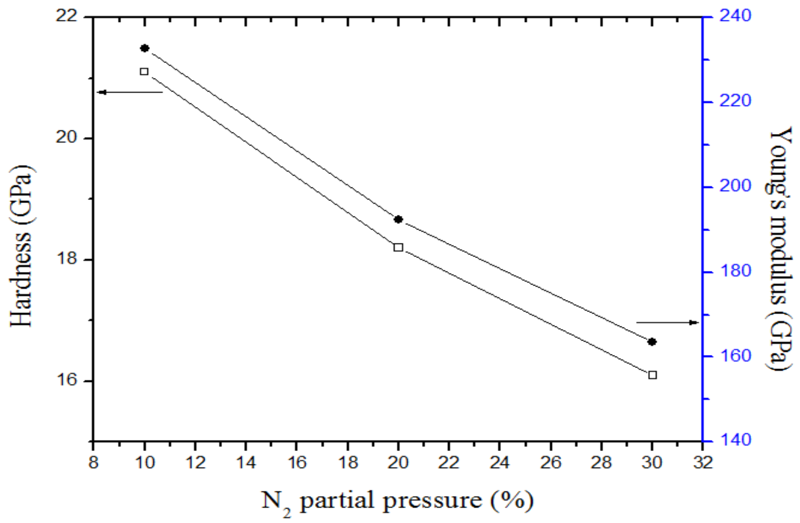

Fig. 3. Hardness and Young's modulus of the TaN films at different partial pressures of N_2.

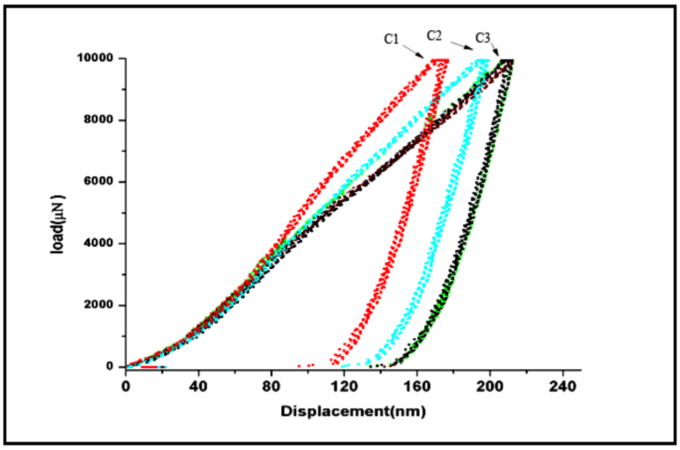

Fig. 4. Load-penetration depth curves of indentations for the TaN films with different N_2 partial pressures.

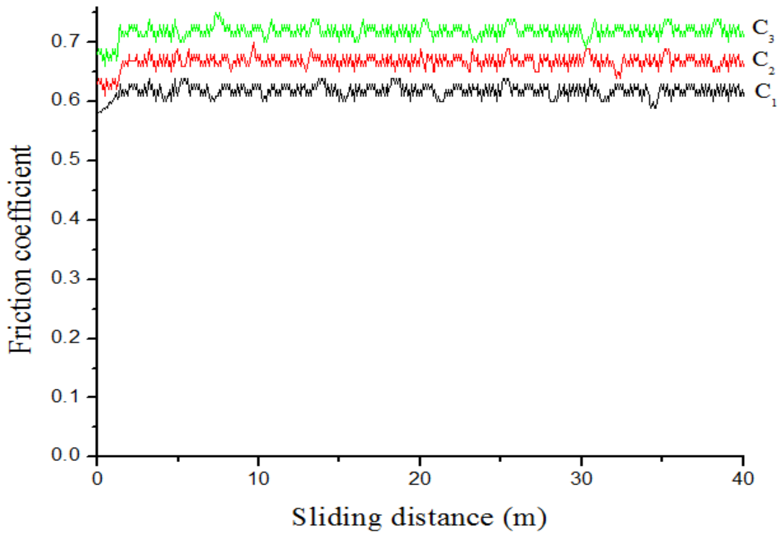

Fig. 5. Friction coefficient as a function of the sliding distance.

The images of various TaN films after wear test are shown in figure 6. Sample C_1 had the narrowest wear track width due to its higher hardness. With increasing N_2 partial pressure, sever tracking and delamination was found along the track, indicating a relatively brittle failure mode. Generally, factors which are effective in improving wear resistance are hardness increase and friction coefficient decrease, because a decrease in the friction coefficient means a decrease of transfer tangent power in the surface of the sample. The images also show that the wear track decreases as N_2 partial pressure decreases, which indicates the improvement of wear resistance.

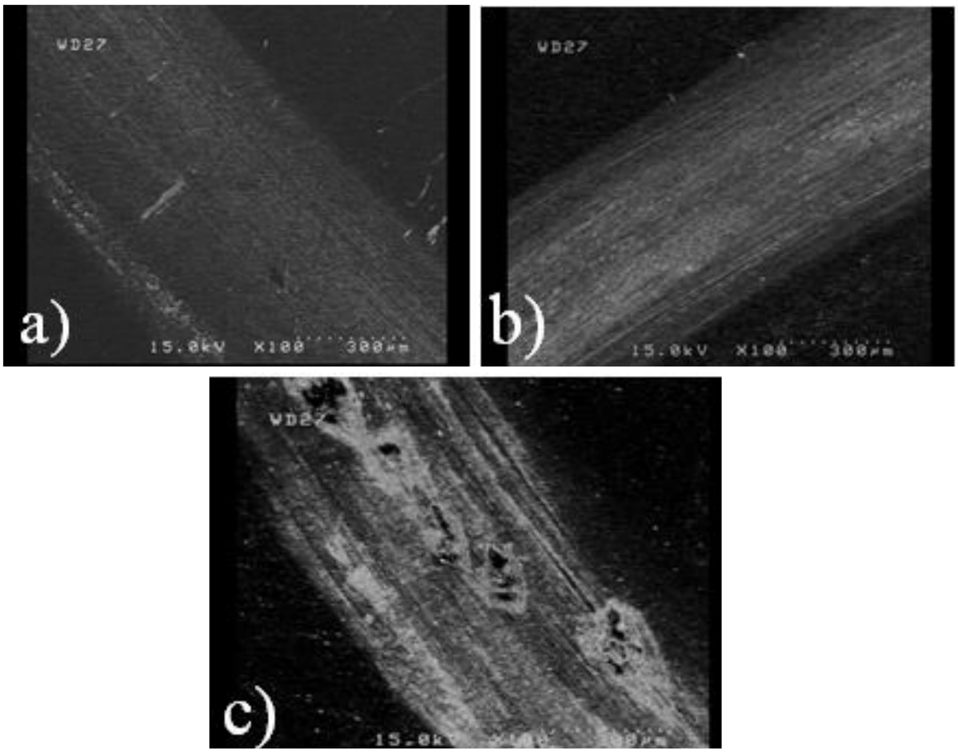

Fig. 6. The images of the formed wear grooves: a) sample C_1, b) sample C_2 and c) sample C_3.

Conclusions

In the present study, the effect of N_2 partial pressure on the microstructure and mechanical properties of TaN films was investigated. The TaN films with crystalline microstructure features were fabricated by magnetron sputtering with Ar:N_2 gas input control of 90:10 %, 80:20 % and 70:30 %, respectively. XRD patterns showed that TaN crystalline phases were formed into (111), (200) and (220) direction with FCC structure at different N_2 partial pressures. According to the AFM images, grain size and roughness increased with increasing N_2 partial pressure. The highest hardness and Young's modulus around 21 and 230 GPa respectively were found for sample C_1 due to its small grain size and roughness. Also, increasing N_2 partial pressure caused the friction coefficient to increase. The images of sample C_1 after wear test showed the narrowest wear track, and with increasing N_2 partial pressure (samples C_2 and C_3) sever cracking and delamination were observed along the wear path.

References

[1] M. Kot, W.A. Rakowski, Ł. Major, R. Major, J. Morgiel, "E ffect of bilayer period on properties of Cr/CrN multilayer coatings produced by laser ablation", Surf. Coat. Technol., Vol. 202, 2008, pp. 3501.
[2] D.G. Liu, C.D. Gu, R. Chen, J.P. Tu," Interface and superhardness of TiN/CN_x multilayer films", Surf. Coat. Technol., Vol. 205, 2010, pp. 2386.

[3] M. Sakurai, T. Toihara, M. Wang, W. Kurpsaka, S. Miyake, “Surface morphology and mechanical properties of nanoscale TiAlN/SiN*x* multilayer coating deposited by reactive magnetron sputtering”, Surf. Coat. Technol., Vol. 203, 2008, pp. 171.
[4] G. Ma, G. Lin, S. Gong, X. Liu, G. Sun, H. Wu, “Mechanical and corrosive characteristics of Ta/TaN multilayer coatings”, Vacuum. Vol. 89, 2013, pp. 244.
[5] H.C. Jiang, C.Wang, W. Zhang, X. Si, Y. Li, “Influences of Film Thickness on the Electrical Properties of TaN_x Thin Films Deposited by Reactive DC Magnetron Sputtering”, J.Mater. Sci. Technol., Vol. 26, 2010, pp. 597.
[6] Y.L. Kuo, J.J. Huang, S.T. Lin, C. Lee, W.H. Lee, “Diffusion barrier properties of sputtered TaNx between Cu and Si using TaN as the target”, Mater. Chem. Phys., Vol. 80, 2003, pp. 690.
[7] Q. Xie, J. Musschoot, C. Detavernier, D. Deduytsche, R.L.V. Meirhaeghe, S.V. den Berghe, Y.L. Jiang, G.P. Ru, B.Z. Li, X.P. Qu, “Diffusion barrier properties of TaN_x films prepared by plasma enhanced atomic layer deposition from PDMAT with N_2 or NH_3 plasma”, Microelectron. Eng., Vol. 85, 2008, pp. 2059.
[8] Y.K. Lee, K.M. Latt, K. Jaehyung, K. Lee, “Study of diffusion barrier properties of ionized metal plasma (IMP) deposited TaN between Cu and SiO_2”,Mater. Sci. Semicond. Process., Vol. 3, 2000, pp. 179.
[9] S.R. Burgess, H. Donohue, K. Buchanan, N. Rimmer, P. Rich, “Evaluation of Ta and TaN-based Cu diffusion barriers deposited by Advanced Hi-Fill (AHF) sputtering onto blanket wafers and high aspect ratio structures”, Microelectron. Eng., Vol. 64, 2002, pp. 307.
[10] S.K. Kim, B.C. Cha, "Deposition of tantalum nitride thin films by D.C. magnetron sputtering", Thin Solid Films., Vol. 475, 2005, pp. 202.