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# Microstructure evolution and protective properties of TaN multilayer coatings

Y.H. Yang, F.B. Wu *

Department of Materials Science and Engineering, National United University, Taiwan

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ABSTRACT

The protective Tantalum nitride (TaN) multilayer coatings staked by the TaN layers with alternatively structures at various deposition input powers were proposed in this study. The TaN multilayer coatings were sequentially deposited with layers at 75 and 150 W by magnetron sputtering. The effects of microstructure and layer thickness for multilayer were investigated. The 20-layer TaN multilayer coating with layer thickness of 50 nm exhibited stratified structure with an amorphous/crystalline altering feature, while the 50-layer one with layer thickness of 20 nm grew with continuous columnar crystalline structures penetrating through interfaces. Higher hardness and Young's modulus were found for the 50-layer TaN multilayer coating approximately 15.1 and 175.5 GPa, respectively. The progressive loading scratch and Rockwell-C adhesion tests were adopted to evaluate the adhesion strength. The anti-corrosion behavior was also analyzed. The amorphous/crystalline structure for the 20-layer TaN multilayer coating showed an elevated critical load for adhesion fracture ($L_{c2}$) around 31.6 N due to the large volume ratios for the amorphous structure at initial deposition stage. Superior corrosion resistance for the 20-layer TaN multilayer coating was found due to its amorphous structure.



## 1. Introduction

Amongst various protective coatings, the multilayer films possess numerous advantages, such as superior hardness, toughness, and corrosion resistance than the single layer coatings. Accordingly, they are widely applied in versatile industries to improve the performance and lifetime for the tool components [1–7]. In general, the multilayer coating is staked by at least two or more materials to combine specific characteristics on demands. However, the use of multiple materials leads to higher costs and complicated processes during manufacturing. In the transition-metal nitride family, tantalum nitride (TaN) systems can form several equilibrium phases as well as metastable structures, which can be modulated by process parameter control to deeply influence the protective properties [8–9]. The tantalum nitride is therefore a potential candidate for developing the hard coating for specific applications by manipulating the structure. Many researchers have demonstrated that the $N_2$/Ar gas inlet ratio is a dominant factor to the microstructure from crystalline to amorphous feature for TaN coating [10–14]. In our previous works, the amorphous TaN layer under the Ar/$N_2$ gas flow ratio of 12/8 sccm/sccm exhibits the merits in anti-corrosion behavior [15] and toughness [16] as compared the crystalline TaN layer under the Ar/$N_2$ gas flow ratio of 18/2. The single element nitride multilayer coating, c-TaN/a-TaN multilayer, has been further developed and shown with better protection performance. On the other hand, the input power control also plays an important role for the microstructure and protective properties. In our latest research [17], the optimal state for the crystalline TaN coating was found under a deposition process parameter of 150 W input power. The TaN multilayer coatings under various deposition powers with 20 and 50 nm layer thickness are successfully manufactured. The TaN multilayer coating with sequentially 75 and 150 W deposited layers showed the superior wear resistance. Nonetheless, the detailed microstructure evolution and other protective properties have not yet been fully understood. Besides, the effects of thickness of the staked layers are also of interest. In this study, 1 μm thick TaN multilayer coatings with total 20 and 50 layers were deposited by reactive magnetron sputtering at 75 and 150 W alternatively modulation. The microstructure, hardness, adhesion, and corrosion resistance of the coatings were analyzed and discussed.

## 2. Experimental procedures

The TaN multilayer coatings were deposited on AISI 420 steel substrate and Si (111) wafer for various analyses. Prior to deposition, all the substrates were cleaned by an ultrasonic agitator with acetone and followed deionized (D.I.) water. The coatings were fabricated by radio frequency (RF) magnetron reactive sputtering. The high purity

* Corresponding author at: A2-344R, No. 2 Lienda Rd., Nan-Shih-Li, Miaoli 36063, Taiwan.
*E-mail address:* fbwu@nuu.edu.tw (F.B. Wu).

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**Table 1**
The deposition conditions of the TaN multilayer coating.

| Coating designation | Input power (W) | Single layer thickness ratio (nm/nm) | Bilayer thickness (nm) | Number of layers |
|---|---|---|---|---|
| M-75/150-20L | 75/150 | 50/50 | 100 | 20 |
| M-75/150-50L | 75/150 | 20/20 | 40 | 50 |

*Gas flow ratio of Ar/$N_2$ was fixed at 18/2.

Ta (99.995%) target with 50.8 and 6.0 mm in diameter and thickness, respectively, was applied as deposition source. The working distance from target to substrate was controlled at 100 mm. The chamber was evacuated down to $3.2 \times 10^{-6}$ Pa and the substrates were kept at 350 °C during deposition. A pre-sputtering process with pure Ar ions under 200 W RF input power for 5 min to clean target surface was performed prior to deposition. A thin interlayer of Ta was firstly deposited under 100 W input power with 20 sccm pure Ar for 10 min to promote the adhesion. After introducing the reactive gas of $N_2$ to a Ar/$N_2$ flow ratio at 18/2 sccm/sccm, the TaN multilayer coatings were fabricated by alternatively sequential stacking of TaN single-layer coatings at 75 and 150 W input powers. The staking layer numbers of 20 and 50 were carefully manipulated for comparison, namely M-75/150-20L and M-75/150-50L, respectively. The overall thickness for TaN multilayer coatings were all controlled around 1 μm, indicating 50- and 20 nm for single layer thickness for M-75/150-20L and M-75/150-50L coatings, respectively. The detailed sputtering conditions for TaN were designated in Table 1. Detailed fabrication procedures can also be found in our previous work [17].

The detailed microstructure observations for the cross-sectional images were carried out through the transmission electron microscopy (TEM, JEM-2100, JEOL, Japan). The selected area electron diffraction (SAED) was employed for the structure feature. The hardness and Young's modulus of the coatings were measured by nano-indentation (TriboIndenter, TI 900, Hysitron, USA). The indentation test was conducted under an applied load of 5 mN with a 10-second dwelling at the maximum load. The measured maximum indentation depth was kept at 80 nm, which was less than one tenth of the coating thickness to avoid the substrate effect. To get accurate mean and standard deviation, all samples were investigated from at least 8 tests and averaged. The Rockwell C adhesion test (Rockwell Hardness Tester, ATK-F1000, Mitutoyo, Japan) using a 0.2 mm radius HRC indenter with an applied

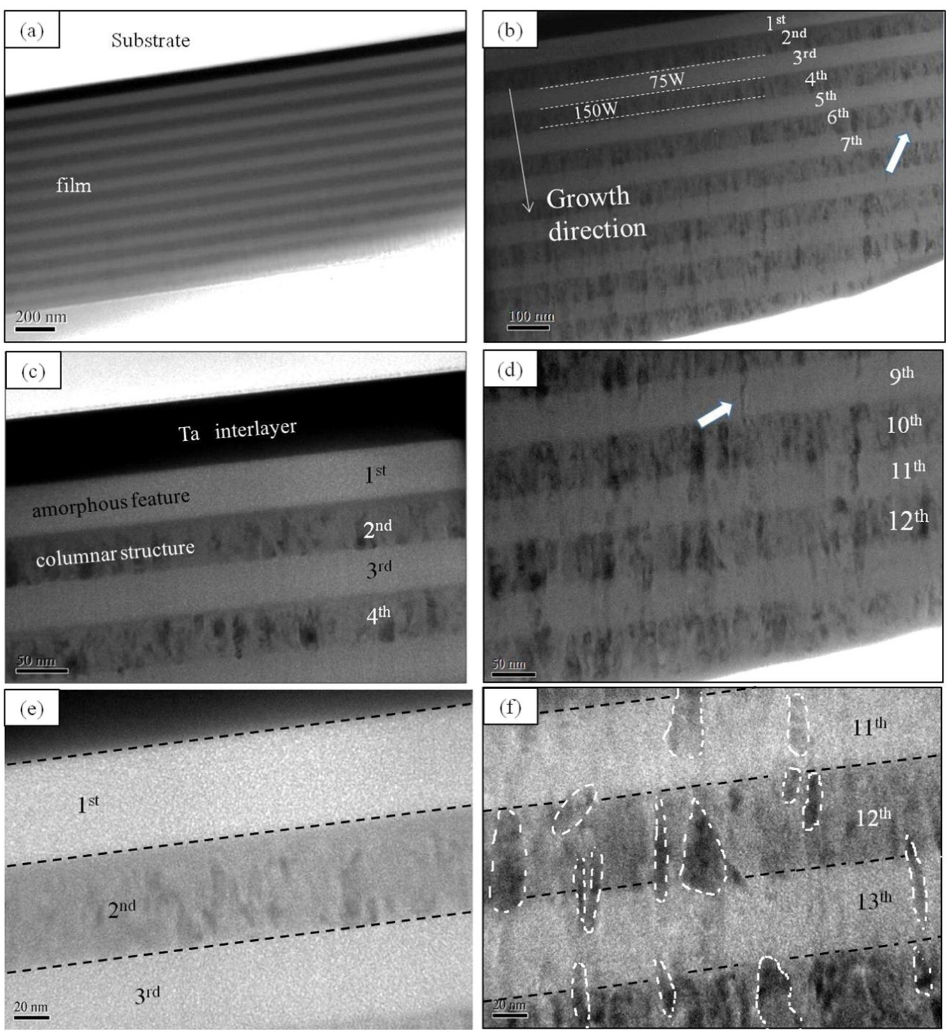


**Fig. 1.** The cross-sectional TEM images of the M-75/150-20L coating at magnification of (a) 10 k, (b) 20 k, (c) (d) 50 k, and (e) (f) 100 k for layered feature.

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