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MD-Simulation of Duty Cycle and TaN Interlayer Effects on the Surface Properties of Ta Coatings Deposited by Pulsed-DC Plasma Assisted Chemical Vapor Deposition

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In this work, molecular dynamics (MD) simulations were employed to investigate the effects of duty cycle changes and utilization of tantalum nitride interlayer on the surface roughness and adhesion of Ta coating deposited by pulsed-DC plasma assisted chemical vapor deposition. To examine the simulation results, some selected deposition conditions were experimentally implemented and characterized through scanning electron microscopy, atomic force microscopy (AFM) and microscratch tests. The Ta and Ta/TaN coatings were deposited on AISI316L stainless steel substrate in a plasma atmosphere consist of Ar, H_2, N_2 and TaCl5 vapor at 350 °C. The results showed that at the same duty cycles the surface roughness of Ta/TaN coating is at least 40% less than that of the single layer Ta coating. By increasing the duty cycle from 17 to 40%, the surface roughness significantly decreases about 80%. This is attributed to the further exposure of surface against high energy ions bombardment at higher duty cycles. The presence of TaN interlayer due to its lower lattice mismatch with Ta (under 2%), contributes to the nucleation of Ta grains which consequently leads to reducing the surface roughness. The enhanced adhesion of the Ta coatings on TaN is discussed in view of improving the interfacial stresses as shown by the MD simulations. The MD simulations results were shown to be in good agreement with the experimental data.

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1. INTRODUCTION

In the last two decades, tantalum coatings have been widely used in energy industries, electrical and micro-electromechanical components as well as biomedical devices owing to their unique properties such as high electrochemical inertness, high melting point (about 3017˚C), high biocompatibility and appropriate strength at high temperatures [1–4]. The Ta thin films exist in two phases: α-Ta which is ductile with BCC crystal structure and β-Ta which is brittle with a tetragonal crystal structure. The adequate ductility of α-Ta in comparison with brittle β-Ta makes it interesting for mechanical applications [5]. It is well understood that utilizing an appropriate deposition technique as well as a compatible interfacial film is necessary to achieve an α- rich Ta coating [6, 7].

Surface roughness and adhesion to the substrate are two most important properties of coatings. These properties remarkably affect film properties. For instance, increasing the surface roughness of Ta coatings typically leads to reduce corrosion resistance [8, 9]. Similarly, low adhesion to substrate results in the coating spallation and considerably decreases its life time [10]. Therefore, minimizing the surface roughness concurrent with maximizing the adhesion to the substrate of Ta-based coatings is very important.

Physical vapor deposition (PVD) and chemical vapor deposition (CVD) processes are two conventional methods

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for the synthesis of Ta-based coatings [7, 8, 11, 12]. PVD techniques such as ion beam assisted deposition and sputtering are known as suitable methods to deposit Ta films at low temperatures with a good deposition rate. However, the serious weaknesses in step coverage and adhesion, have constrained them to develop upto the industrial scale. In last few years, the plasma assisted chemical vapor deposition (PACVD) has been introduced as a method of choice for producing high quality films with favorable adhesion and uniform surface coverage at relatively low temperatures (below 450˚C). In this method, due to the use of plasma, the thermodynamical limitations of conventional CVD techniques are eliminated and the deposition reactions take place at relatively low temperatures [10]. PACVD is a well-established deposition method from an industrial standpoint since it has the advantages of PVD techniques, alongside its capability in coating the components with complex geometries [13].

It is well accepted that the substrate's chemical composition, grain size and surface topography as well as deposition parameters can affect the roughness and adhesion of deposited coatings. The final properties of the deposited film must be considered taking into account the simultaneous effects of process parameters. However, due to the limitations of the experimental conditions, the simultaneous controlling and understanding of these parameters are too difficult. In this regard, molecular dynamics (MD) simulations are known as a favorable technique to overcome the abovementioned limitations [14]. The microstructural properties of atomic layers such as Ta-based coatings can be effectively examined using MD simulations.

In the scope of the microstructural properties simulations of Ta films, Nikravesh et al. [15], reported the surface tribology examination of Ta thin films through MD simulations that had been deposited via magnetron sputtering technique. They found that application of a TaN interlayer on stainless steel substrate prior to deposition of the Ta coating has a positive effect on reducing the roughness and grain size of sputtered Ta thin films about 78% and 34%, respectively. They also indicated that the higher sputtering powers (incident energy) and lower temperatures were led to the formation of films with smoother surfaces. In another work, Firouzabadi et al. [16] numerically investigated the sputtering power effects on the nano-tribological properties of TaN films using MD simulation. They indicated that increasing sputtering power from 30 to 60W resulted in increasing the deposition rate from 1.88 to 3.26 m/h. Also, by increasing sputtering power from 1.5 to 3 W/cm^2, the surface roughness reduced to a minimum value of 0.5 nm and then increased to 1 nm. They numerically showed that this condition is associated with decreasing incident-atom energy at higher deposition rates and there is an optimized value of deposition rate and incident-atom energy which give a minimum surface roughness.

The optimal conditions for the deposition of α-rich tantalum coatings by pulsed DC PACVD are previously studied by Ghorbani et al. [17]. It is well accepted that the PACVD is a new, high quality and cost-effective approach to produce tantalum coatings, but it still has a lot of unknown aspects. Among various PACVD parameters, the duty cycle plays a key role as a kinetic and thermodynamic variable which can directly affect the ionization process. So, this paper attempts to uncover the actual interplay between changing some parameters in PACVD of Ta and some key characteristics of the resulting Ta layer through a combination of MD simulations and experimental verifications. More precisely, in the present study, MD simulations are conducted to comprehensively investigate the effects of duty cycle and TaN interlayer on the tribological properties, film adhesion and interfacial stress of Ta films deposited on AISI 316L stainless steel (SS) by pulsed-DC PACVD technique. Furthermore, to validate the results of MD simulations, the surface roughness and the adhesion of deposited coatings were experimentally examined using an atomic force microscope (AFM) and microscratch techniques, respectively. The roughening mechanisms during film growth and how the TaN interlayer affects the surface roughness and adhesion strength of the deposited Ta coatings are discussed in detail.

2. EXPERIMENTAL PROCEDURE

2. 1. Simulation Method MD simulations were conducted to explore the effects of utilizing TaN interlayer on surface roughness, adhesion to substrate and also growth models of Ta coating on the SS substrates under different duty cycles. The movements of all mobile atoms and substrate atoms were ran by the second law of Newton. They were calculated by the direct integration from classical Hamiltonian equations of the motions using Velocity-Verlet technique. The deposition procedures were simulated by the constant micro-canonical ensemble (NVE) with constant number of particles (N), the volume of system (V) and the system's total energy (E). Each Ta atom entered the simulating box from random points, x and y coordinates, affected by the atomic collisions and finally received at a point on surface with varied velocity and kinetic energy. In the pulsed-DC PACVD, the deposition reaction takes place efficiently only when pulsed is on. Therefore, in this simulation, the tantalum atoms entered the system at different time intervals, according to the duty cycle.

In MD simulations, the Morse potential is used for showing the interaction between both substrate and Ta atoms. The Morse potential can be written as [18]:

$$U_{rj} = D\left[e^{-2\alpha(r_{ij}-r_0)} - 2e^{-\alpha(r_{ij}-r_0)}\right] \quad (1)$$

where U is an energy function of pair potential, r_{ij} the distance between atoms i and j, r_0 the nearest atomic distance at equilibrium, D the cohesion energy and α a fitted parameter for material which corresponds to the material binding tension energy and the bulk modulus. In the Morse function, due to the different interactions of atomic species, more parameters are required. These parameters can be determined using the Lorentz–Berteloth mixing lows using the following equations [18]:

$$D_{\text{A-B}} = (D_A D_B)^{0.5} \quad (2)$$

$$\alpha_{A-B} = 0.5(\alpha_A + \alpha_B) \quad (3)$$

$$r_{0A-B} = (\sigma_A \sigma_B)^{0.5} + ln\left\{\frac{2}{\alpha_{A-B}}\right\} \quad (4)$$

$$\sigma_{A,B} = r_{0A,B} - ln\left\{\frac{2}{\alpha_{A-B}}\right\} \quad (5)$$

The applied Morse parameters of this work are reported in Table 1 [16, 19].

In order to increase both the accuracies of results and computational efficiency, the computational time step was set to be 1 fs[1] on the substrates with the dimensions of 15×60×215 Å.

The MD simulations were carried out using the large-scale atomic/molecular massively parallel simulator (LAMMPS) [20] and the post analyses were imagined by OVITO [21]. MD simulations can be used to study the surface diffusion, morphology, growth regime and interfacial stress of coatings to study the simultaneous effects of different deposition parameters.

The energies of incoming species were assumed as their kinetic energy which is related to their velocity. After MD simulations, the root means square (RMS) surface roughness of the samples were determined using the following equation [22]:

$$RMS = \left(\frac{\sum_{i=1}^{N}(Z-\overline{Z})^2}{N}\right)^{0.5} \quad (6)$$

where the index i indicates all atoms along the X (or Y) direction, Z is the mean surface position in the surface normal, z, direction and N is the total number of atoms. The rate of surface diffusion of tantalum atoms and interfacial stress between coatings and substrate were determined. The average horizontal displacement of Ta atoms was considered as the base of measuring the interface diffusion rate. The stresses were characterized by a formula that calculates the symmetric per-atom stress tensor for each atom in a group. The tensor of each atom has 6 components and is considered as a 6 elements vector in the following order: xx, yy, zz, xy, xz, yz. A virial contribution generated by a small set of atoms is attributed in equal portions to each atom in the set [23]. It should be mentioned that, before performing the MD simulations of deposition processes, the physicochemical properties of SS substrate and TaN interlayer had been introduced to software based on the elements data, crystal structures and lattice constants of each material.

TABLE 1. The values of Morse parameters of each pair used in MD simulations

Substance	D (eV)	α (Å^{-1})	r_0 (Å)	σ (Å)
Ta-Ta	0.7504	1.1319	3.346	2.8793
N-N	0.2	6.087	1.8316	2.2946
Fe-Fe	0.4216	1.3765	2.849	2.3823
Ta-N	0.3872	3.6094	2.0503	-
Fe-N	0.2903	3.7317	1.7948	-
Fe-Ta	0.5623	1.2542	3.0857	-

2. 2. Experimental Verification

The SS substrates with the dimensions of 20 mm×20 mm×2 mm were mechanically grounded by #1000-#5000 SiC sandpapers and polished with 0.1 µm Al_2O_3 suspension to mirror finishing. After that, an ultrasonic cleaning process was conducted in acetone to remove all residual contaminants. The prepared substrates were then set on the holder (cathode) of pulsed-DC PACVD system. The reaction chamber was a cylinder with a diameter and length of 50 and 70 cm, respectively. The chamber wall was the anode and it was heated by thermal elements. The temperature was monitored using thermometers. Before starting the PACVD Process, sputter etching was performed to remove the residual contaminants from the substrate's surface. The deposition parameters which were applied in this study of pulsed DC-PACVD, are shown in Table 2.

Surface topography and coatings roughness were analyzed using an atomic force microscope (AFM, Araresearch Company in contact mode). The cross-sectional morphologies of deposited coatings were studied

[1] Femtosecond equals to 10^{-15} seconds

TABLE 2. The process conditions for pulse DC-PACVD of Ta coatings on 316L stainless steel at 350

	Temperature (°C)	P (torr)	Gas flow ratio (%)			Deposition time (min)	F (kHz)	I (A)
			H_2	Ar	N_2			
Sputter etch	350	11	20	75	15	20	8	4
Tantalum monolayer	350	5	60	40	0	60	11	2.5
TaN interlayer	350	5	50	25	25	30	11	2.5

using field emission scanning electron microscopy equipped with an EDS line scan analysis module (FESEM, TESCAN MIRA3). The adhesion of deposited layers was investigated by a microscratch test apparatus equipped with a Rockwell-C indenter and optical microscope (OM). A normal load from 0.1 to 50 N was increasingly applied along a 10 mm distance on coated samples.

3. RESULTS AND DISCUSSION

The cross-sectional images generated using MD simulations of the Ta coatings achieved at some selected duty cycles on the SS substrate with and without TaN interlayer are indicated in Figure 1.

It is noticeable that with increasing duty cycle and using TaN interlayer, the roughness of the deposited Ta layer decreases. The effects of duty cycle and use of TaN interlayer on the surface profile of the deposited Ta coatings are represented in Figure 2.

The RMS surface roughness values of the deposited coatings were directly determined using surface profiles extracted from MD simulations. Figure 3 shows the normalized RMS surface roughness values obtained in various duty cycles with and without TaN interlayer. By dividing each value by the maximum value, the normalized values can be achieved.

Considering Figure 3, it can be seen that the normalized surface roughness values of the deposited Ta coatings on TaN interlayer are less than those of the coatings deposited at the same duty cycles on SS substrate. Also, with increasing duty cycle from 15 to 40%, the normalized surface roughness value for the coatings deposited directly on SS substrate decreases from 0.88 to 0.33 and in the presence of TaN interlayer it decreases from 0.57 to 0.27.

In order to verify the MD simulation results, the deposition of Ta on the SS substrate and TaN interlayer was conducted at some duty cycles. The surface topography of samples was investigated by AFM. The results are shown in Figure 4. From this figure, it can be observed that increasing duty cycle and using TaN interlayer contribute to the formation of smoother coatings.

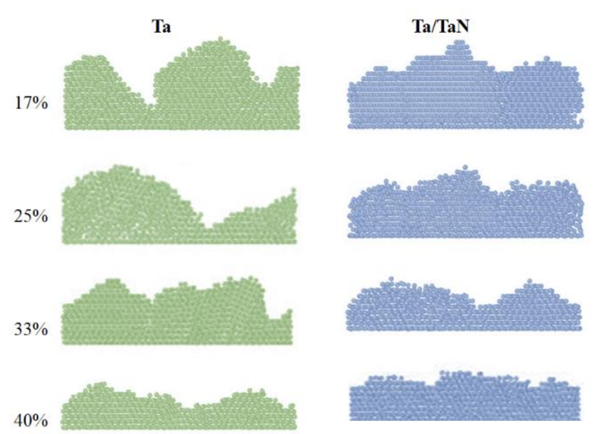

Figure 1. The cross-sectional images generated using MD simulations of the Ta coatings at some selected duty cycles on (a) SS surface and (b) TaN interlayer

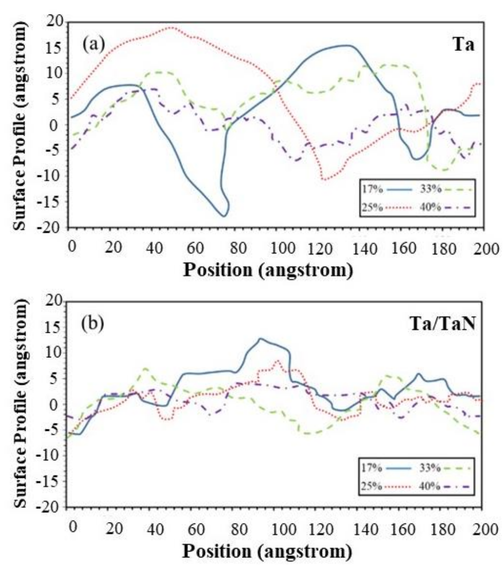

Figure 2. Simulation results of the cross-sectional profiles of the surface for Ta and Ta/TaN systems at various duty cycles

Figure 5 shows the effects of the interlayer and duty cycle on the RMS surface roughness and grain size of experimentally deposited coatings. The effects of TaN interlayer on the changes of RMS surface roughness and grain size are clear. Using TaN interlayer leads to reducing both of these parameters.

From Figure 5, it can be concluded that the effects of TaN on decreasing RMS surface roughness are more dominant. Based on the results of Figure 4 and Figure 5, it seems that using TaN interlayer and also deposition at higher duty cycles leads to formation of smoother

Ta coatings. In other words, the deposited Ta coatings. Moreover deposition at higher duty cycles corresponds to more exposure of surface under discharge bombardment. In this conditions a huge number of atoms can be sputtered from the surface. This not only results in the formation of a smooth surface, but also tends to reduce the deposition rate.

The cross-sectional SEM images of the Ta coatings deposited at various duty cycles are represented in Figure 6. It seems that the reduction of coatings thickness is attributed to two phenomena. At first, it is well-known that in pulsed-DC deposition growth mechanisms take place predominately in pulse-off times while production of Ta atoms and solid phase nucleation prevails when pulse is on. In this regard, an optimum pulse-off time is essential for the Ta atoms to easily diffuse along the surface and receive the preferred positions in the lattice structure [24]. In this condition, production of a dense and flawless film is expected. Moreover at higher duty cycles, surface is under a longer ion bombardment which can detach the weak adhered atoms or clusters from the coatings on TaN interlayer are smoother than those of the deposited on the SS substrate. This can be attributed to the nucleation effect of TaN. According to the results of similar works [25–27], the presence of nitrogen can provide preferential sites for

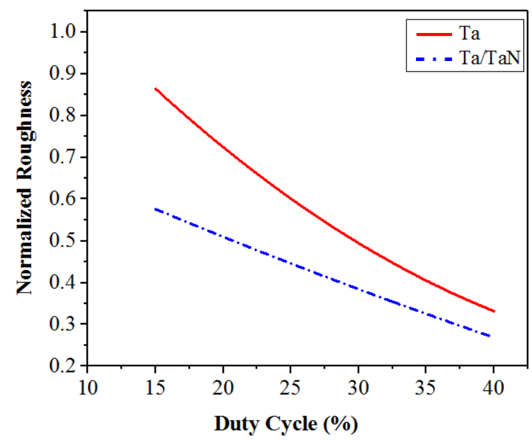

Figure 3. Normalized surface roughness values determined by MD simulation of the deposited Ta films as a function of duty cycle with and without TaN interlayer

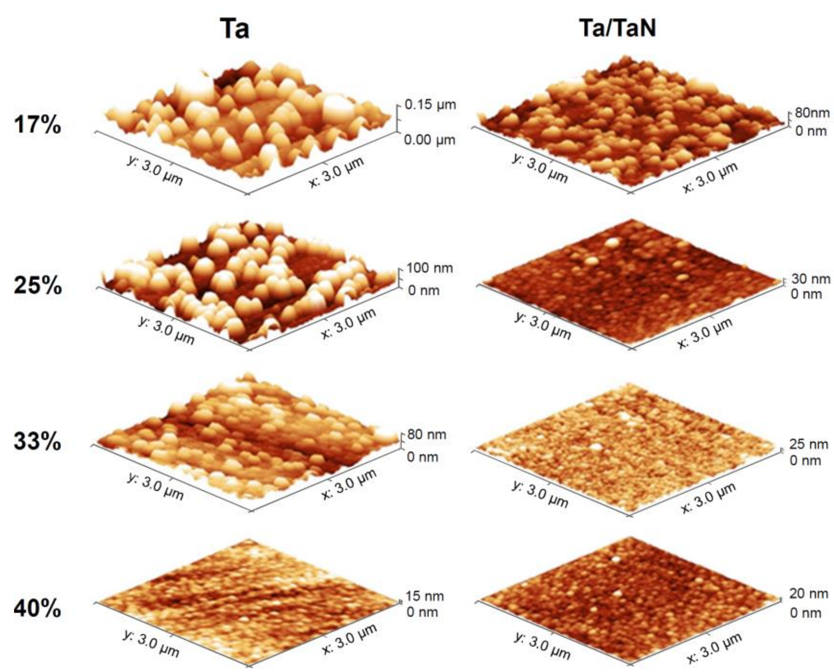

Figure 4. Surface topographical images of the Ta coatings deposited on SS substrate and TaN interlayer achieved by AFM

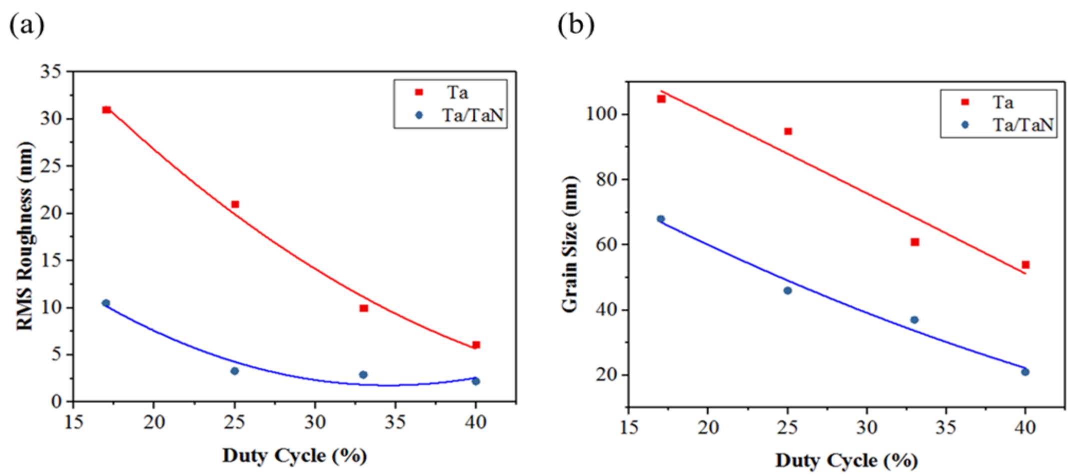

Figure 5. (a) RMS surface roughness values and (b) grain size of deposited Ta coatings with different duty cycles on SS substrate and TaN interlayer, obtained from AFM results

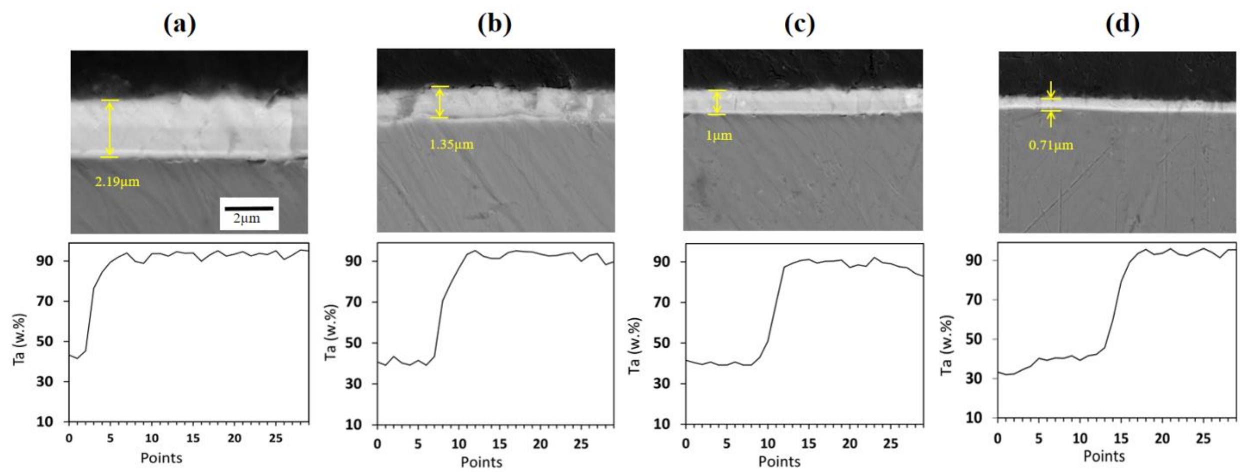

Figure 6. Cross-sectional SEM Images and EDS line scan chemical analysis of the Tantalum coatings deposited on 316L SS at different duty cycles of (a) 17%, (b) 25%, (c) 33% and (d) 40%

nucleation of subsequent Ta grains on the surface. Consequently, increasing the nucleation rate can prevent grain growth leading to produce smoother surface. Secondly, the production of more Ta atoms at higher duty cycles (more pulse on times) results inthe reduction of each's incident energy which causes to stop them before reaching the steady state positions in lattice. These atoms can easily send out from the surface under ion bombardment. Therefore, the simultaneous effects of these two phenomena increases the number of atoms which are potential to detach from the surface under ion bombardment especially at higher duty cycles. These observations are in accordance with the results of S. Konstantinidis et al. [24] who showed that by increasing the duty cycle in the PECVD of alumina films, their surface topography being rougher and both the deposition rate and deposited mass decrease. They also found that ion bombardment induced desorption, densification and sputtering of deposited films are the consequences of the increased pulse-on time (duty cycle).

The other important feature of a coating is its adhesion to substrate. Figure 7 illustrates the variations of normalized interfacial stress values with different duty cycles of the deposited Ta films on both the SS substrate and TaN interlayer calculated from MD simulations. It is evident that in both coatings, with increasing duty cycle,

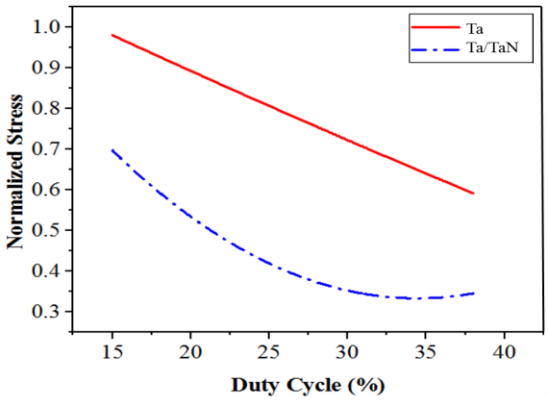

Figure 7. Normalized interfacial stress values obtained from MD simulations as a function of duty cycle with and without TaN interlayer

the interfacial stress between the deposited Ta layer and substrate is decreased. This phenomenon can be attributed to the excess energy that enters to surface at higher duty cycles. The additional thermal energy corresponds to rising the surface temperature and enhances the diffusion of Ta atoms. In this condition, the Ta atoms fill the lattice imperfections easily and lattice defects considerably decline. This results in lower interfacial stress between coating and substrate. However, it seems that the presence of TaN interlayer has a promising effect on the reduction of interfacial stress. Apparently, the lower amounts of calculated interfacial stress in Ta/TaN bilayer system are more associated with the lower lattice mismatch between Ta and TaN which has previously reported by Gladczuk et al. [28]. Therefore, the higher adhesion of Ta coating on TaN interlayer can be attributed to a higher level of crystalline match, and hence, the reduction of interfacial stress between coating and substrate.

To validate the results of the MD simulations about the influence of TaN interlayer on interfacial stress, the microscratch tests were carried out. Figure 8 (a) and (b) show the OM images of the microscratch tests tracks of Ta/SS and Ta/TaN samples. The failure models observed in this test are also shown in Figure 8(c). It can be seen that the Ta coating deposited on the SS substrate is detached with the formation of Hertzian cracks. It is well-known that Hertzian cracks generally, form in the coatings with lower adhesion to the substrate [10, 29]. From the abovementioned discussion, it can be concluded that the higher amount of interfacial stress exists at the interface of Ta coating and SS substrate. However, in the case of Ta/TaN system, some small spallation with negligible amounts of conformal cracks imply to an adherent and uniform coating. It is worth noting that this condition generally takes place in the coatings with less interfacial stress. Thus, considering the lower crystalline mismatch between Ta and TaN [28] and also the results of microscratch tests, it can be concluded that the interfacial stress in Ta/TaN system is much lower than that of Ta/SS substrate.

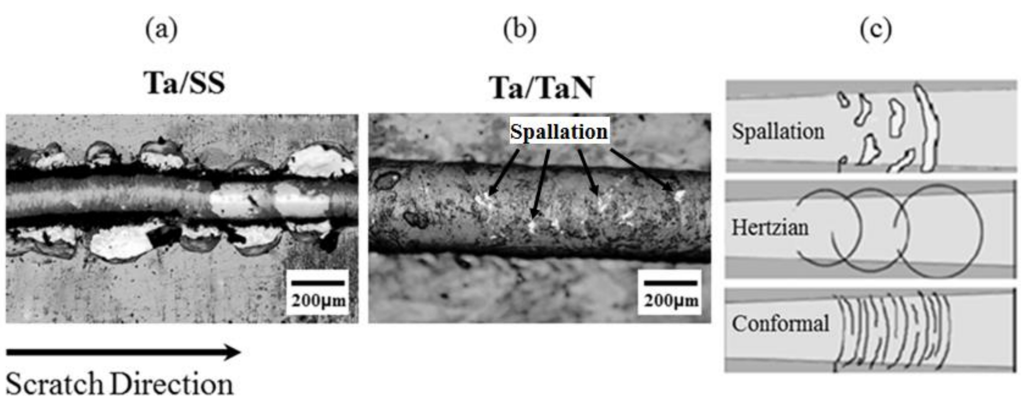

Figure 8. Tracks of the diamond tip after micro scratch tests on samples of (a) Ta films deposited on bare SS substrate and (b) Ta films deposited on TaN interlayer and (c) the schematic illustration of coating failure models after scratch test.

4. CONCLUSIONS

In this work, the effects of TaN interlayer and duty cycle of PACVD on the surface roughness and adhesion strength of Ta coating deposited on AISI 316L stainless steel were investigated, via MD simulations and experimental examinations. The main conclusions are as follows:

1. The results of MD simulations are consistent with the experimental examinations including AFM and microscratch tests for both the deposited Ta coatings on

SS substrate and the TaN interlayer under the same conditions.

2. At the duty cycle of 40% (pulse-on times), the sputtering of atoms from the surface of coatings was intensified. This resulted in the reduction of films surface roughness and thickness of about 80% and 67%, respectively.
3. The deposition of Ta on TaN interlayer resulted in the formation of films with smooth surfaces. This is attributed to more preferential nucleation sites of Ta on TaN in comparison with bare SS substrate.
4. The better film adhesion in the Ta/TaN system can be attributed to its lower interfacial stress as suggested by the results of MD simulations.

5. ACKNOWLEDGMENT

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Persian Abstract

چکیده

در این تحقیق، به‌منظور بررسی اثرات تغییرات چرخه کاری و نیز استفاده از میان‌لایه نیترید تانتالوم بر زبری سطح و چسبندگی پوشش تانتالوم که با روش رسوب شیمیایی از فاز بخار به کمک پلاسما (PACVD) ایجاد شده بود؛ از شبیه‌سازی دینامیک مولکولی (MD) استفاده شد. برای ارزیابی نتایج شبیه‌سازی، برخی از شرایط رسوب انتخاب شده به‌صورت تجربی اجرا و بوسیله میکروسکوپ الکترونی روبشی(SEM) ، میکروسکوپ نیروی اتمی (AFM) و آزمون خراش مشخصه‌یابی شد. پوشش های Ta و Ta/TaN روی زیرلایه فولاد زنگ‌نزن AISI316L در یک محیط پلاسما متشکل از Ar ، H_2 ، N_2 و بخار پنتاکلراید تانتالوم ($TaCl_5$) در دمای ۳۵۰ درجه سانتیگراد ایجاد شدند. نتایج نشان داد که در چرخه‌های کاری یکسان، زبری سطح پوشش Ta/TaN حداقل ٪۴۰ کمتر از زبری سطح پوشش تک‌لایه Ta است. با افزایش چرخه کاری از ٪۱۷ به ٪۴۰ ، زبری سطح، به طور قابل توجهی، در حدود ٪۸۰ کاهش می‌یابد. این کاهش زبری می‌تواند بدین علت باشد که با افزایش چرخه کاری، زمان قرارگیری سطح در معرض بمباران یونهای پرانرژی افزایش می‌یابد. وجود میان‌لایه TaN به‌واسطه فاکتور عدم انطباق کریستالی کمتر آن با تانتالوم (کمتر از ٪۲) منجر به افزایش نرخ جوانه‌زنی دانه های Ta شده و نهایتاً سبب کاهش زبری سطح می‌شود. همانطور که توسط شبیه‌سازی MD نیز نشان داده شد؛ چسبندگی بیشتر پوشش‌های Ta روی میان‌لایه TaN، ناشی از اصلاح میزان تنش در فصل مشترک می‌باشد. نشان داده شد که نتایج شبیه‌سازی MD تطابق خوبی با داده‌های تجربی دارد.