Tribology International 105 (2017) 185–192

Contents lists available at ScienceDirect

Tribology International

journal homepage: www.elsevier.com/locate/triboint

A comprehensive study on the surface tribology of Ta thin film using molecular dynamics simulation: The effect of TaN interlayer, power and temperature

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ARTICLE INFO

Keywords:
Molecular dynamics
Roughness
Sputtering
Tantalum
AFM characterization

ABSTRACT

Molecular dynamics simulations (MD) was carried out to investigate the surface nano-tribology of grown Ta sputtered thin films on stainless steel with and without TaN interlayer. Ta and TaN films were deposited by magnetron sputtering technique at different temperatures. Argon and nitrogen mixture gas was used to create TaN interlayer. Using AFM technique, the surface of sputtered thin films was characterized. MD simulation results were validated by experimental results and a good consistency was observed the two techniques. The effects of TaN interlayer and process parameters were investigated. The results showed that the surface roughness can be remarkably improved by using a TaN seed layer. Higher sputtering powers at lower temperatures provide the best condition to produce smoother Ta films.

1. Introduction

Recently tantalum based films have been extensively studied and used owing to their excellent properties such as corrosion resistance, high conductivity, biocompatibility, high melting point, high ductility and high strength at high temperatures [1–5]. They can potentially be applied in proton exchange membrane fuel cells (PEMFCs), biocompatible coating in vitro and vivo [5,6]. Ta thin films are currently used in semiconductor industry for microelectronic devices [7–9] and mechanically protecting coatings [10,11]. However, Ta films, which are deposited via sputtering, have not the same properties (such as surface roughness, porosity, grain size and etc.) as the bulk material due to differences in their processing techniques and structures.

Surface roughness of deposited thin films is practically one of the most important factor in bio-compliant coatings on medical implants [12,13], friction coefficient of coating [14,15], the interfacial contact resistance (ICR) in PEMFCs system [16], adhesive strength of nano-scale contacts [17] and components for water wetting properties [18]. Furthermore, as reported by Maeng [19] and Evgeny [20], the increase of surface roughness of deposited films may lead to increase of corrosion rate which significantly affects both protein adsorption and cellular response in biomedical applications [12]. Indeed, reducing surface roughness is generally interested in the fabrication of thin films [21].

Magnetron sputtering is a well-established process to produce tantalum films based on physical vapor deposition (PVD). The surface roughness of deposited films by magnetron sputtering is, therefore, affected by the process parameters such as glancing angle of deposition [22], pressure and power [23], temperature [24,25] and so on. Due to the limitations of experimental conditions, it is difficult to understand and control simultaneously all affecting process parameters. However, molecular dynamics (MD) simulations is an interesting method which helps to overcome abovementioned limitations. MD simulations is a powerful tool for examining the atomic scale structure of deposited atomic layers [26] which has been used for first time in tantalum sputtering process with TaN seed layer.

In the present work, MD simulations of Ta deposited thin films were carried out onto both AISI 316L (SS) substrate and TaN seed layer under different deposition process conditions. Actual sputtered Ta thin films were also applied onto similar surfaces under the same conditions, by magnetron DC sputtering technique, to validate MD simulations results. The effects of power, temperature and TaN seed layer were studied on the roughness of the resulted sputtered thin films. In order to identify the affecting parameters on the roughness variations, MD simulations were employed to calculate surface atom diffusion rate and interfacial stress under various conditions.

2. Simulation method

Molecular dynamics simulation was carried out to investigate the

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http://dx.doi.org/10.1016/j.triboint.2016.10.010
Received 29 August 2016; Received in revised form 7 October 2016; Accepted 8 October 2016
Available online 11 October 2016
0301-679X/

effect of tantalum nitride seed layer on growth mechanism of tantalum thin film on the AISI 316L stainless steel (SS) substrate under different sputtering powers and substrate temperatures. Fig. 1 shows a three-dimensional model of Ta atoms depositing onto a substrate with the dimensions of 140*20*20 Å. The motions of all mobile adatoms and substrate atoms are governed by the Newton's second law. They are determined by the direct integration of the classical Hamiltonian equations of motion using Velocity-Verlet method. The deposition process is simulated using constant micro-canonical ensemble (NVE) with constant number of particles (N), the system's volume (V) and the total energy in the system (E). Each depositing tantalum atom enters the simulating box from a random points, x and y coordinates, and it will rest at a point on surface with minimum energy.

In MD simulation, the Morse potential is applied for expressing the interaction between both substrate atoms and adatoms. The Morse potential is written as [27]:

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

Where U is a pair potential energy function, r_{ij} is the distance between atoms i and j, r_0 is the nearest atomic distance at equilibrium, D is the cohesion energy and α is a material fitted parameter that depends on the binding tension energy of the material and the bulk modulus. For the different atomic species interaction, more parameters are needed in the Morse function. These parameters can be calculated using the Lorentz–Berteloth mixing rules using the following equations [28]:

$$D_{A-B} = (D_A D_B)^{1/2} \quad (2)$$

$$\alpha_{A-B} = \frac{1}{2}(\alpha_A + \alpha_B) \quad (3)$$

$$r_{0A-B} = (\sigma_A \sigma_B)^{1/2} + \ln\left(\frac{2}{\alpha_{A-B}}\right) \quad (4)$$

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

The Morse parameters used in this research are listed in Table 1 [28,29]:

Considering both computational efficiency and accuracy of results, the computational time step was set 1 fs. 3600 Ta atoms were deposited during simulation runs. The substrate temperatures were set from 25 to 300 °C and the incident energies were set from about 0.08–2 eV.

The simulation processes were performed using the large-scale atomic/molecular massively parallel simulator (LAMMPS) [30] and the post analyses were visualized by OVITO [17,31].

MD simulation method can be systematically performed to analyze the growth process, morphology, surface diffusion and interfacial stress of the deposited thin films in details in order to study the simultaneous effects of processing parameters.

After the simulation process completed, the mean surface roughness of the samples, *RMS*, were calculated using Eq. (6) [32,33].

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

where the index i runs over all atoms along the X (or Y) direction. N is the total number of atoms, while $\overline{Z}$ is the mean surface position in the surface normal, z, direction.

The surface diffusion rate of adatoms and interfacial stress between sputtered films and substrate were calculated. The average horizontal displacement of Ta adatoms was the base of measuring the interface diffusion rate.

The stresses were defined by a computation that computes the symmetric per-atom stress tensor for each atom in a group. The tensor for each atom has 6 components and is stored as a 6-element vector in the following order: xx, yy, zz, xy, xz, yz. A virial contribution produced by a small set of atoms is assigned in equal portions to each atom in the set [34].

It is required to mention that, before the simulation process of sputtering, substrate (stainless steel and TaN) had been constructed, using the information of elements, crystal structures and lattice constants related to the desired structure.

3. Materials and method

In the present work, discs of AISI 316 L stainless steel (SS) with dimensions of 30 mm in diameter and 8 mm in thickness were used as substrates. The disc surfaces were polished with different grades of polishing papers to obtain smooth mirror like surfaces. The polished discs were subsequently cleaned in acetone and methanol ultrasonic bath to remove any surface contamination.

Ta was deposited by magnetron sputtering with Ta target 99.95%. Argon with the purity of 99.99% was used as plasma gas and nitrogen with the purity of 99.99% was used as reactive gas to create TaN sputtered seed layer. Sputtering processes were applied under base pressure of 5×10^{-6} Torr and the working pressure of 5×10^{-3} Torr. Tantalum thin films were deposited with and without TaN interlayer under different sputtering conditions of temperature and power as presented in Table 2:

Intermediate sputtered films were applied on some samples, as shown in Table 2, by nitrogen leaks into the sputtering chamber before final Ta film deposition. After applying the interlayers, with interruption of nitrogen flow without breaking vacuum, Ta sputtering operation stage was performed.

Morphological investigations and roughness measurements were performed using atomic force microscope (AFM) VEECO and subsequently the prepared images were analyzed by Gwyddion software.

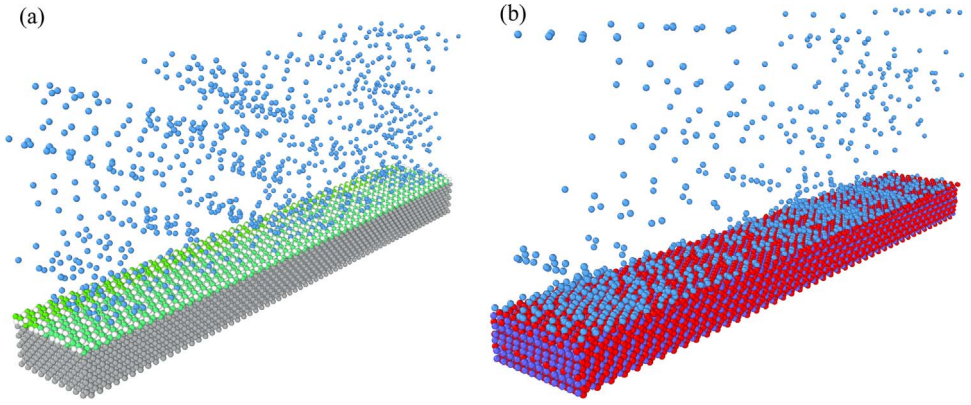

Fig. 1. Simulation model of the deposition process, (a) on stainless steel and (b) on TaN interlayer; (Gray: Fe in SS substrate; White: O in Cr2O3; Green: Cr in Cr2O3; Light blue: Ta Adatoms; Red: N in TaN and Blue: Ta in TaN). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)