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Evaluation of residual stress in sputtered tantalum thin-film

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ABSTRACT

The influence of deposition conditions on the residual stress of sputtered tantalum thin-film has been evaluated in the present study. Films have been deposited by DC magnetron sputtering and curvature measurement method has been employed to calculate the residual stress of the films. Transitions of tantalum film stress from compressive to tensile state have been observed as the sputtering pressure increases. Also, the effect of annealing process at temperature range of 90–300 °C in oxygen ambient on the residual stress of the films has been studied. The results demonstrate that the residual stress of the films that have been deposited at lower sputtering pressure has become more compressive when annealed at 300 °C. Furthermore, the impact of exposure to atmospheric ambient on the tantalum film stress has been investigated by monitoring the variation of the residual stress of both annealed and unannealed films over time. The as-deposited films have been exposed to pure Argon energy bombardment and as result, a high compressive stress has been developed in the films.

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1. Introduction

Tantalum (Ta), a refractory metal, has a wide range of applications in a variety of fields. Due to its high melting point (3290 K), high density, high fracture toughness and corrosion resistance, tantalum is a promising material in bio-medical, micro electromechanical system (MEMS) devices and high-temperature applications [1]. Tantalum thin-films have been playing an important role in electronic devices acting as a diffusion barrier between metals and silicon substrate [2–4], as an alternative of hazardous electrodeposited chromium coatings [5] and an excellent x-ray mask absorber [6]. Tantalum is used also as a hardener coater on titanium surface for orthopaedic implants applications [7] and to protect steel and glass substances from wear and corrosion [8,9]. Recently, tantalum has been employed successfully for fabricating buckled and straight MEMS beams [10] and as a metal bridge structure for the resonant gate transistors (RGTs) that could be vibrated at low range of frequency suitable for audio applications [11].

However, one of the main obstacles that may lead to functional failure in a device is the residual stress of the material. For instance, buckling and cracking in films, wafer curvature and changes in device functions could be attributed to undesirable stress effects [12,13]. Despite numerous attempts to characterise the stress in thin-film materials, the real mechanism is not well understood. Several approaches have been reported to control the residual stress or improve the physical properties of tantalum thin-film for a specific application. It has been found that the residual stress and crystallographic structure of tantalum thin-film can be manipulated in various ways, such as annealing at different temperatures, using different substrates and deposition techniques and changing the deposition conditions. Previous work have been published about the influence of annealing process, thermal cycling and oxygen diffusion on the intrinsic stress and phase transition between alpha (α) and beta (β) of tantalum thin-films [14–21]. The stresses of tantalum films that have been deposited on different substrates including silicon [14,15], silicon dioxide [14,17], stainless steel [5,22], glass [13,23], and titanium [24] have been investigated. Several studies have been reported on the stress of tantalum thin-films as a function of substrate bias voltage [6,22], deposition system [6,20,22,23,25], deposition conditions [10,20,23,26–29], substrate temperature [6], ion bombardment [30] and film thickness [5,12].

However, previous studies have limited their research on a particular aspect of deposition or annealing conditions and the conditions' influence on the phase transformation of the tantalum films. In our study, we aim to provide a comprehensive investigation of the tantalum film stress as a function of deposition conditions, annealing treatment, exposure of annealed and unannealed film to atmospheric ambient and ion bombardment exposure. The present work utilises the curvature method to deter-

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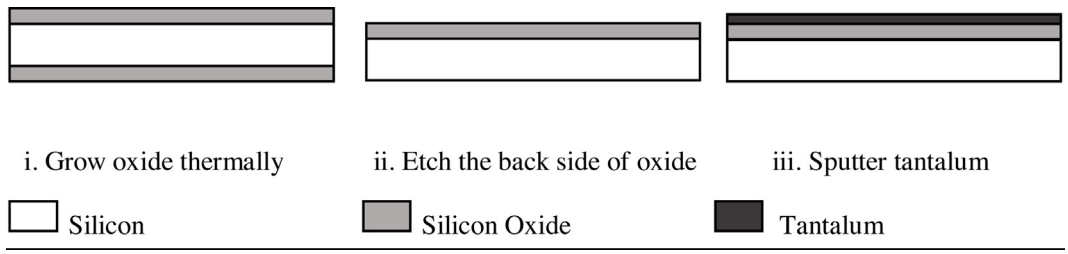

Fig. 1. Fabrication process flow for tantalum thin-film.

Table 1
Sputtering parameters of tantalum thin-films in DC magnetron system using argon (Ar) as sputtering gas with flow rate of 50 sccm.

Wafer	Power (W)	Pressure (mTorr)
1	300	3.5
2	300	7.5
3	300	11.5
4	300	19
5	300	24
6	400	3.5
7	400	7.5
8	400	11.5
9	400	19
10	400	24
11	500	3.5
12	500	7.5
13	500	11.5
14	500	19
15	500	24

mine the total residual stress, including: intrinsic stress that arises from the microstructure and deposition process, thermal stress that occurs due to the thermal coefficient differences, and extrinsic stress that arises from post-deposition processes. Importantly, the main objective of this study is to identify the conditions of sputtering, annealing and air or ion bombardment exposures under which the residual stress will behave in a more compressive or tensile manner. The anticipated outcomes of these results are very valuable and could be employed in the fabrication of MEMS devices for sensing and actuation applications.

2. Experiment details

Fig. 1 shows the fabrication steps of tantalum thin-film. To begin with, about 0.5 μm thick oxide has been grown thermally on both sides of a silicon substrate (4-inch, <100> p-type). After removing the backside of oxide via reactive ion etching (RIE), an initial set of curvature measurement has been performed using (Veeco Dektak 8000 Surface Profiler). Then, a set of 50 nm tantalum thin-films have been deposited on the top oxide by DC magnetron sputtering system (OPT Plasmalab system 4000). The sputtering conditions are presented in Table 1.

In this process, a step has been created which enables film thickness to be measured by atomic force microscopy (AFM) and Dektak profilometer. Afterwards, a second set of curvature measurement has been carried out and the residual stress $\left(\sigma^f\right)$ of the films has been estimated based on Stoney's formula:

$$\sigma^f = \frac{E_s T_s^2}{6(1-\upsilon_s)T_f}\left(\frac{1}{R_{s+f}} - \frac{1}{R_0}\right) \quad (1)$$

where E_s is the modulus of the substrate (MPa), υ_s is Poisson's ratio of the substrate, T_s is the substrate thickness (μm), T_f is the film thickness (μm), R_0 is the initial substrate radius of curvature (μm) and R_{s+f} is the final radius of curvature (μm) of substrate plus the thin-film.

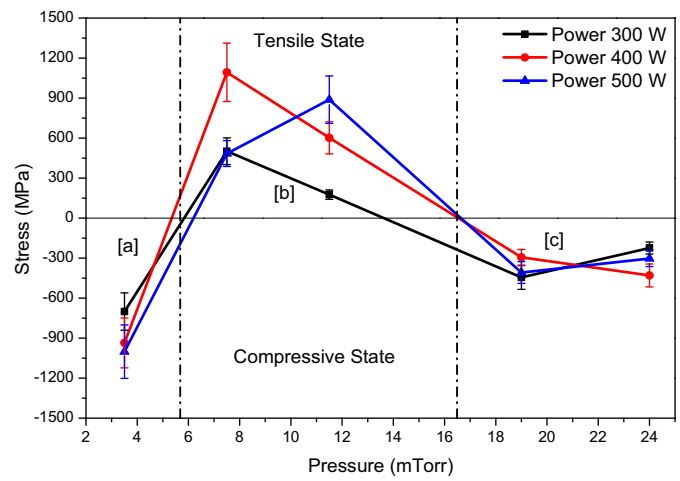

Fig. 2. The residual stress of tantalum films as a function of sputtering pressure for different sputtering powers.

3. Results and discussion

3.1. Effect of sputtering pressure and power

The residual stress measurements of as-deposited tantalum films as a function of sputtering pressure for powers (300, 400 and 500 W) are shown in Fig. 2. Three main regions of the residual stress in the films are shown in the figure: compressive stress (regions a and c) and tensile stress (region b). It can be seen that the effect of sputtering power on the degree of residual stress in the films have been found to be significantly larger in the tensile state region. In the compressive stress regions, increasing the power has produced a slight change in the residual stress of the films. Consistent with literature findings [10,20,26,27,31], the residual stress begins in a compressive state (region a) at low pressure (3.5 mTorr) and as the sputtering pressure increases, the trend of stress switches to a tensile state (region b) at 7.5 and 11.5 mTorr before shifting back again to the compressive state (region c) at 19 and 24 mTorr respectively.

The compressive stress exhibited in tantalum films at low sputtering pressure (region a) could be because of the atomic peening. The atomic peening mechanism has been described in more detail elsewhere [23,28,32]. In such a scenario, the argon and target atoms have long mean free path and high momentum with fewer collisions, thus bringing about a compressive stress in the film. Therefore, the compressive-stressed film at low sputtering pressure might have dense fibrous structures as described by Yoshihara and Suzuki [33].

In the second region (b), the stress of tantalum films is observed to change from compressive to tensile around 7.5 mTorr. It seems that the tensile-stressed films could possess ultra-small columnar grains with low density grain boundaries at the film/substrate interface [20], and this abrupt transition could be attributed to constrained grain boundary relaxation [23,28,32]. In

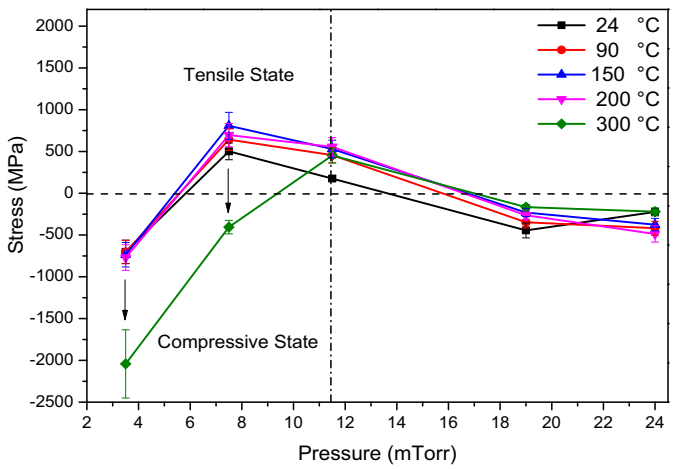

Fig. 3. The residual stress as a function of sputtering pressure for the annealed tantalum films at different temperatures.

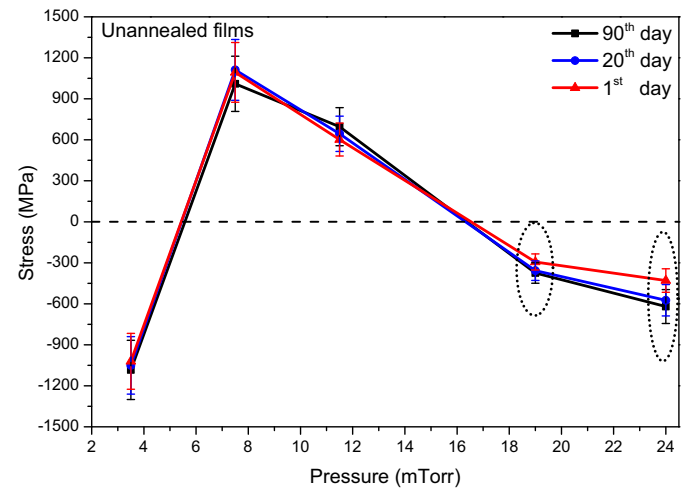

Fig. 4. The residual stress as a function of sputtering pressure for the different exposure times to atmosphere for the unannealed films.

addition, this transition in the stress between compressive and tensile could be associated to phase transformation between α and β as reported previously [16,26,29]. For instance, Navid and Hodge [26] have found that the α phase has been developed in the compressive region, whereas β or mixed phases have been developed in the tensile state. The third region of transition from tensile to compressive stress at higher sputtering pressures 19 and 24 mTorr could be related to the mechanism of impurity incorporation, where oxygen based contaminants are desorbed from the deposition chamber [27,32]. In general, at high deposition pressure (from 11.5 to 24 mTorr), energy and flux of particles are decreased because of the increase of collisions with non-directional ion bombardment. Consequently, columnar grains and porous microstructures are developed.

3.2. Effect of annealing in oxygen ambient

Fig. 3 depicts the measured stress as a function of sputtering pressure for the annealed films. Firstly, the stress measurement of the as-deposited films has been carried out at room temperature (24 °C). Next, the sputtered films have been subjected to annealing process at temperatures (90, 150, 200 and 300 °C) in oxygen ambient. The stress measurements of the annealed films have been performed directly after each annealing step.

It can be seen that the impact of annealing on the change in the residual stress is largest when the tantalum films have been deposited at lower pressure (from 3.5 to 11.5 mTorr) and annealed at a temperature of 300 °C. The transition towards the compressive stress at low sputtering pressures could be attributed to the formation of tantalum pentoxide Ta_2O_5 layer at the film surface because of the ability of oxygen content to penetrate easily into the unit cell of the films during annealing [18–20,27]. It is possible that the amount of Ta_2O_5 increases as the annealing temperature increases and therefore increasing the compressive stress [18]. In the case of the films that have been deposited at higher sputtering pressure (19 and 24 mTorr), the stress tends to be less compressive after annealing treatment at 300 °C. These influences could be related to the annealed columnar structures that prevent the oxygen diffusion from penetrating easily into the grain boundaries [33]. However, many published work have revealed different interpretations on the relationship between stress shifting and phase transformation. Possible phase transformation may take place at high annealing temperatures up to 600 °C [14,20]. Since the tantalum films in the present study have been sputtered on SiO_2/Si substrate, the oxygen content in the SiO_2 could prevent any kind of phase transformation at a temperature of only 300 °C.

3.3. Effect of atmospheric exposure

Further investigation has been carried out to examine the impact of atmospheric conditions on the residual stress of both annealed and unannealed films. In the case of the unannealed films, three main measurements of stress have been carried out in cleanroom ambient at 24 °C; after deposition directly, after 20 days and after 90 days. Fig. 4 shows the measured stress as a function of sputtering pressure for different exposure time of the unannealed films.

For all exposure periods, a modest change has been observed in the residual stress of the unannealed films that have been deposited at low sputtering pressure. However, the films that have been deposited at higher pressures, 19 and 24 mTorr, exhibit a noticeable increase in the compressive stress after 90 days of the exposure. In the case of the annealed films (at 300 °C only), the stress measurements have been conducted in cleanroom ambient at 24 °C after the first and the 70th day of annealing process. Fig. 5 depicts the measured stress as a function of sputtering pressure for different

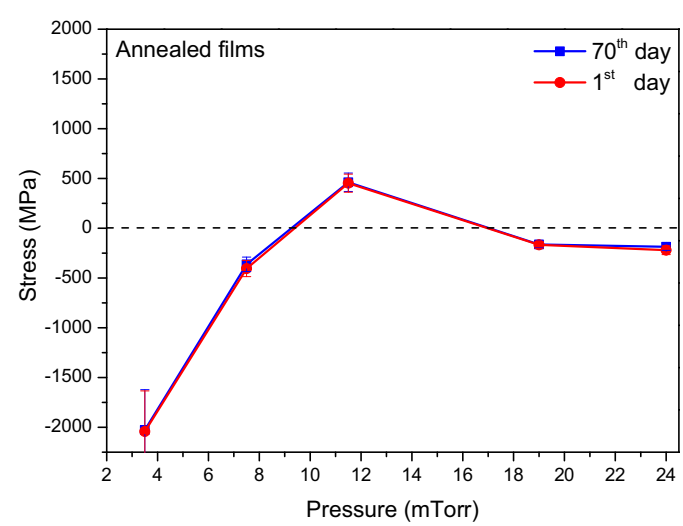

Fig. 5. The residual stress as a function of sputtering pressure for the different exposure times to atmosphere for the annealed films.

exposure time of the annealed films. Indeed, no significant change has been observed in the films after 70 days of the exposure. In this way, no matter how long the films have been exposed, the residual stress can be minimised or remained stable after annealing as reported [6,10,33].

Our results agree with the general trend of stress that has been reported by Yoshihara and Suzuki [33], where the as-deposited films at low (high) pressure might have fibrous (columnar) structures. As a result, the fibrous structures in the unannealed films could hinder the oxygen diffusion from penetrating freely into the grain boundaries as a function of time. On the other hand, the columnar structures in the unannealed films could allow a quick diffusion along the grain boundaries of tantalum films and show a distinguishable increase in the compressive stress when exposed to atmosphere for a long time. It may appear that the annealing process renders both the fibrous and the columnar structures less permeable to oxygen diffusion as a function of time.

3.4. Effect of argon energy exposure

Stress relaxation of tantalum thin-films has been investigated by employing inert-gas (Ar) ion bombardment. Fig. 6 compares the stress measurements as a function of sputtering pressure for unexposed (as-deposited) and exposed films to different ion bombardment parameters. In general, all exposure conditions exhibit a change towards the compressive stress region. By using a power of 100 W and a flow rate of 20 sccm, the residual stress has become two times more compressive, Fig. 6a.

From Fig. 6b, it can be seen that doubling the power to 200 W has changed the residual stress to the compressive state significantly. By increasing the flow rate, the compressive stress has seen to increase further, as indicated in Fig. 6c. The results found in this work could be related to the degree of ion bombardment of the tantalum film, whereby the atoms of material surface are affected by the energy and the amount of bombardment [30,34].

4. Conclusion

In this work, the relationship of the sputtering conditions, annealing treatment, oxygen exposure, and bombardment exposure to the residual stress of tantalum thin-film have been studied. 50 nm of tantalum thin-films have been deposited by DC magnetron sputtering system at different deposition parameters including pressure of (3.5, 7.5, 11.5, 19 and 24 mTorr) and power of (300, 400 and 500 W) respectively. The results reveal that the residual stress of the films starts as a compressive stress at low sputtering pressures and switches significantly to the tensile state as the pressure increases. At higher sputtering pressures, the residual stress has changed from the tensile state to the compressive state. Annealing process has been carried out to investigate the impact of annealing temperature (90, 150, 200 and 300 °C) on the residual stress of tantalum films. The findings show that annealing at 300 °C has produced the most shift to compressive stress in the films that have been deposited at lower sputtering pressure. In addition, noticeable changes of stress in the unannealed films that have been sputtered at higher sputtering pressure have been observed after exposing the films to atmospheric ambient. In contrast, the annealed films have shown a stability in its stress during exposure to the air. Furthermore, the effect of Ar energy exposure has caused the films to become more compressive when the power and flow rate of Ar energy increase. The outcome of this research could be successfully employed for a number of MEMS-based device applications.

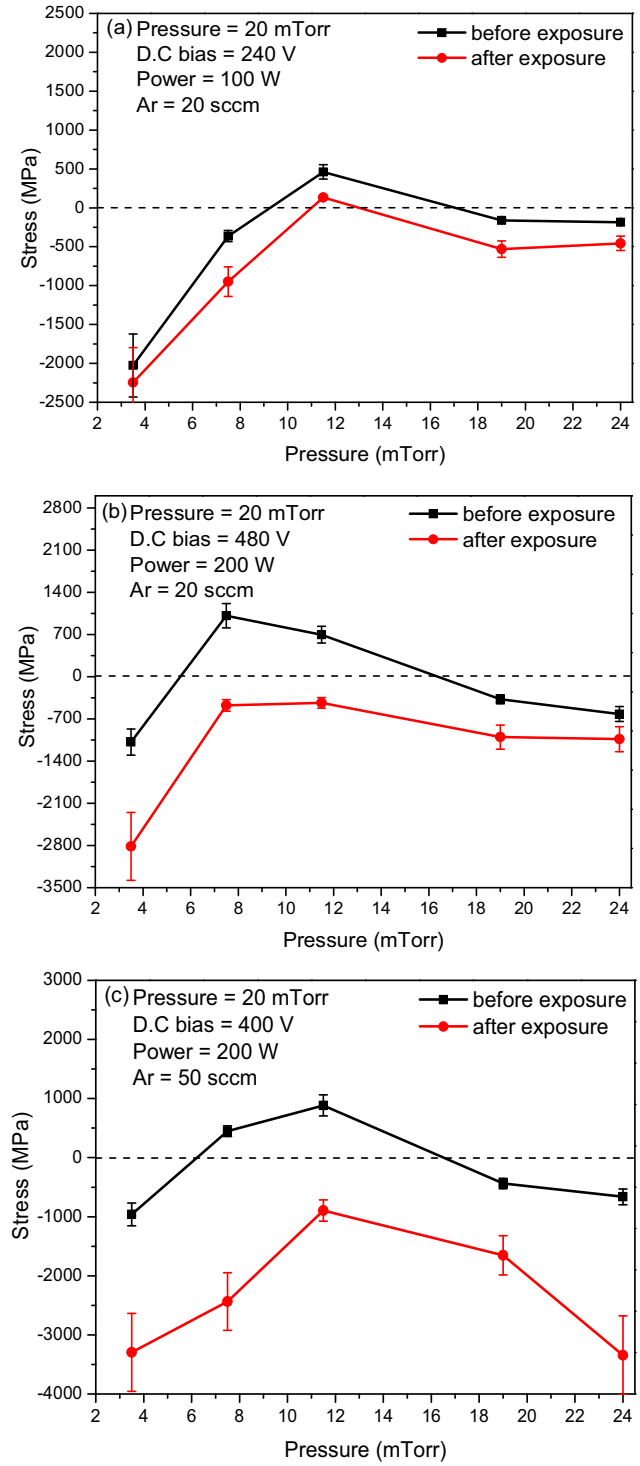

Fig. 6. Comparsion of the residual stress as a function of sputtering pressure for the different conditions of exposure to ion bombardment.

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