High rate and low loss Ta_2O_5 thin films deposited by novel remote plasma reactive sputtering

Y. K. BU, Z. LIU, L. J. CHEN, Z. P. CAI, N. CHEN*
Department of Electronic Engineering, Xiamen University, Xiamen, Fujian 361005, China

In this work we report a novel remote plasma deposition technology for high-rate DC reactive sputtering deposition of low loss Ta_2O_5 films. Deposition rate and optical properties were mainly investigated with process conditions change. This technique is based on generating an intensive plasma remotely from the target and magnetically steering plasma to the target to realize sputtering deposition. It overcomes inherent limitations such as racetrack formation on target and low rate deposition in conventional sputtering technology and realizes fully uniform erosion over the target. Dense Ta_2O_5 thin films with a high deposition rate 0.53nm/s, excellent optical properties are obtained.

(Received July 1, 2010; accepted August 12, 2010)

Keywords: Plasma source, Sputtering deposition, Ta_2O_5 thin films

1. Introduction

Ta_2O_5 thin films have high dielectric constant, high refractive index in the visible spectral region and a wide transmittance spectra region from 0.35 μm-10 μm. They have received mostly attention as capacitor materials [1], and as a high refractive index material for interference coatings [2, 3]. Ta_2O_5 thin films are also investigated for high temperature resistance due to their excellent chemical stability. Deposition of Ta_2O_5 thin films has been accomplished using different deposition techniques such as: electron beam evaporation [4], ion-assisted deposition [5], reactive magnetron sputtering [6] and ion beam sputtering [7]. Various properties of the films characteristics have been reported [8-10].

The films by sputtering deposition have high packing density and low optical absorption and scatter losses. Conventional reactive sputtering deposition normally has very low deposition and high compressive stress. Especially in magnetron sputtering, because of racetrack formation over the surface of target, this has a strong effect on the process instabilities and rate drifts. Generally for metal-oxide thin films deposition by DC reactive magnetron sputtering, a metal target is used for achieving a high deposition rate, but this often leads to local target poisoning, low deposition rate and process instabilities.

The remote high density plasma source was developed in 2001 by Prof. Mike [11]. This technique overcomes inherent limitations of conventional sputtering and has been used successfully in the fields of flexible display and ferromagnetic materials [12, 13]. In this paper, the process was developed as a method of depositing high refractive index, low loss Ta_2O_5 thin films with a high deposition rate.

2. The remote plasma sputtering

A schematic drawing of the system is shown in Fig. 1. Unlike conventional magnetron sputtering, the technique relies on the generation of a plasma remotely from the targets, the internal magnetic elements behind the target are eliminated. An RF coil antennae surrounding a quartz tube is attached to the vacuum chamber as a side arm. The plasma is initiated and amplified by a DC launch electromagnet towards the plasma source exit. This is visible as a color change in the glow discharge from pink to blue (as shown in Fig. 2). A DC steering electromagnet is used to focus and control the direction of the plasma. By tuning the current of the launch and steering electromagnets, the plasma beam is directed to cover the full surface area of the target (as shown in Fig. 3). Obvious difference from conventional sputtering is that plasma density don't depend on the target bias voltage, mainly being set by the remote plasma RF power and system operating pressure. But the plasma energy is low, by applying a DC negative bias (0-800V) to the target, the argon ions are accelerated into the target resulting in a high energy plasma over the full surface area of the target. Hence, fully uniform erosion over the surface of the target is realized and results in a significant reduction in target poisoning. Process instabilities and deposition rate drifts in magnetron sputtering are eliminated because of no racetrack formation on the target. The target material is ejected at a uniform rate across the target surface. This allows for a uniform reaction in the plasma phase when performing reactive sputtering, leading to the formation and deposition of material with a uniform stoichiometry.

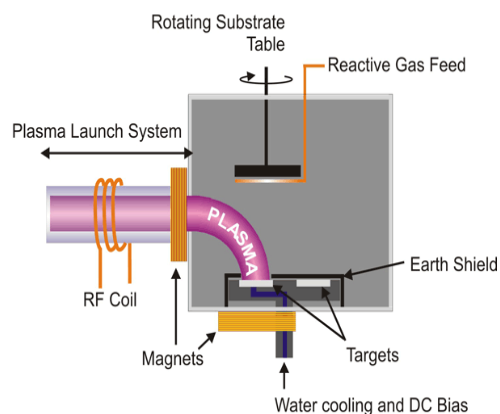

Fig. 1. Schematic drawing of remote plasma system.

The target, mounted on a water-cooled holder, was a high pure metal tantalum (99.99%) disk 100 mm in diameter, 6mm thick. Pure Argon was introduced to the chamber by placing a diffusion ring close to the target, the oxygen was fed into the chamber though another diffusion ring placed as close as possible to the substrate. This was done to maintain the target in a "metal mode" we were able to deposit good quality metal-oxide in the "high growth rate" region of the hysterisis curve, far from the runaway point.

Fig. 2. Photo of remote plasma source.

Fig. 3. Photo of plasma beam deflexion running.

During the deposition process, the system was pumped using cryo-pump with a base pressure of 6×10^{-6} Torr; The oxygen flow was regulated from 0-30 sccm, the argon flow was fixed at 85sccm, and the background pressure is approximately 5×10^{-3}Torr level. To achieve a high deposition rate in reactive process, the RF power was set to 2.2 kW and the target negative bias voltage was set to 700 V respectively. The BK7 glass with 30mm diameter, 3 mm thick, and high surface quality 10-5 scratch dig was used to deposit the films. The deposition time was 10minutes for every single layer. Transmission measurements were performed using Perkin-Elmer Lambda 750 spectrometer. The optical constants, n and k, were calculated from the transmission spectra by the envelope method [14], which is based on the analysis of the transmission spectra of weakly absorbing film that deposited on an free-absorption substrate. Meanwhile the deposition rate was measured and monitored by the Telemark 880 quartz crystal thickness control. Ta_2O_5 thin films were deposited by DC reactive remote plasma sputtering. Main work focused on determining the optimal reactive process transparency window for low loss Ta_2O_5 films. Here deposition rate and optical properties of Ta_2O_5 thin films were mainly investigated with process conditions change.

3. Results and discussion

The deposition rate observed is dependent on the oxygen flow as shown in Fig. 4. The deposition rate initially increases and then decreases with increasing oxygen flow. In initial state, oxygen is zero flow, the deposited film is pure metal Ta film, the rate is keep in a stable value 0.32 nm/s; Then deposition rate begins to increase from 0.32 nm/s to 0.55 nm/s with increasing oxygen flow from 0 sccm-12.5 sccm; Then the deposition rate keeps in a relative stable state with oxygen flow changing from 12.5-15.5 sccm and has no obvious change; Continuing to increase oxygen from 16sccm to 20 sccm, the deposition rate drops sharply and becomes almost constant 0.06nm/s over 24 sccm; However, too much oxygen (more than 30 sccm) lead to runaway process on the target. So the whole run process with oxygen increasing can be divided into four area: metal mode, sub oxide mode, full oxide mode, runaway.

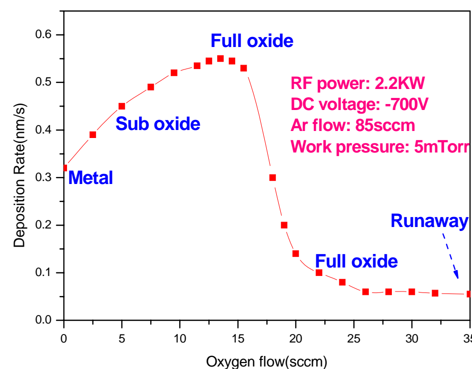

Fig. 4. Deposition rate as a function of oxygen flow.

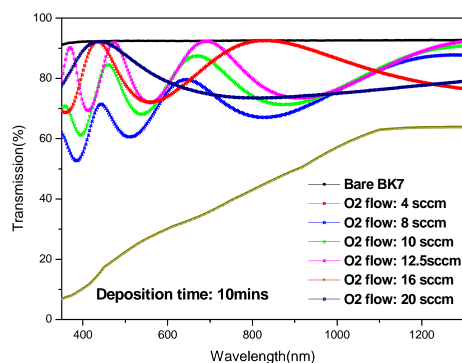

Fig. 5. Transmission spectra of Ta_2O_5 films at different oxygen flow.

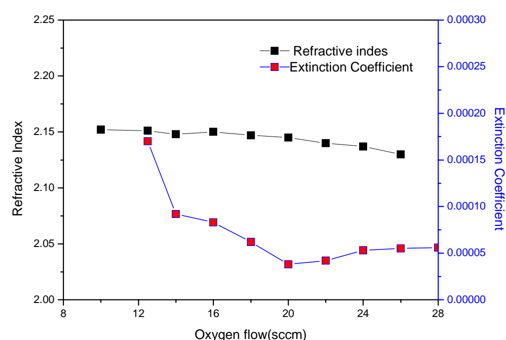

Fig. 6. Refractive inex and extinction coeffcient at wavelength 510 nm with oxygen flow changing.

To investigate the optical properties of Ta_2O_5 films, the transmission spectra were measured with the change of oxygen flow, as shown in Fig. 5. Corresponding to change of deposition rate, the transmission of single layer increased as the oxygen increasing, and the film color changed from the dark brown to full transparency. All the single layers have the same deposition time, 10 minutes. From the transmission spectra we could see the different deposition thickness because of the different deposition rate. Oxygen flow 12.5 sccm is the threshold flow, Over the threshold flow, the spectra transmission at half-wave point reaches the highest value which is equal to the bare BK7 substrate, and this means that at this condition the deposited film has the lowest absorption loss, stoichiometric ratio films Ta_2O_5 were obtained. Beyond the full oxidation threshold(as assessed by optical transmittance) deposition rate decreased with increasing oxygen input to the process, but there was no obvious change in the film refractive index n, the optical constants of Ta_2O_5 films at wavelength of 510 nm with increasing oxygen flow in the full oxide window are plotted in Fig. 6. While the extinction coefficient was dependent on oxygen flow. The extinction coefficient value was 1.83×10^{-4} to 5.1×10^{-5}, as oxygen flow varied from 12.5sccm to 28sccm. The lowest extinction coefficient 3.85×10^{-5} was obtained at oxygen flow of 20 sccm. The extinction coefficient increased a little when oxygen flow increased from 22sccm to 28 sccm. Fig. 7 shows the dispersion data for Ta_2O_5 films at oxygen flow of 15 sccm with a high deposition rate 0.53 nm/s and comparing with conventional reported values. The refractive index at 510 nm for Ta_2O_5 is 2.152, it's significantly larger than EB or IAD because of high energy density plasma impact.

Based on the deposition rate and optical constants change of Ta_2O_5 films, we can see the increase of deposition rate at the initial stage could be volume expansion occuring when the initial metal Ta is converted into sub-oxide TaO and then into the full-oxide Ta_2O_5. The decrease of deposition rate is derived from the serious surface oxidization of the tantalum target at high oxygen flow, and the sputtering yield of the compound is much less than that for the metal. The reduction in deposition rate is the characteristic property for many elements reactively sputtered in Ar/O2 mixtures when the transition from the metal mode to the oxide mode occurs [15]. In addition, the mean free distance of sputtering atom starts to drop when the pressure in the chamber is more than a certain value. This leads likewise to a decrease in the deposition rate. After an insulating film forms on the target surface, it can charge up. Although this can be avoided by pulsed DC power at a certain extent, too much oxygen (more than 30 sccm) lead to arc discharge process, i. e. runaway process.

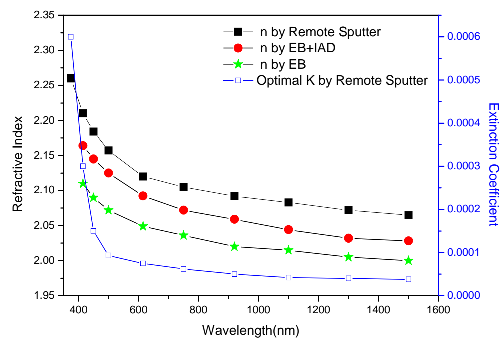

Fig. 7. Dispersion curve for Ta_2O_5 films prepared at a oxygen flow of 15sccm.

For high rate and low loss oxide films, the oxygen flow is very key parameter. The amount of oxygen should not be too high since the unneeded oxygen will be trapped into the film and make voids in it. These voids are very

easy to cause scattering when light pass through the film. The too much oxygen will also cause the obvious drop in deposition rate, even runaway process. On the other side, the amount of oxygen should not be too low because the oxide deficiency causes the optical absorption.

The novel conformation of remote plasma sputtering provides a very stable reactive sputtering process at a high deposition rate. During reactive oxidation in the remote plasma process, very high density plasma contains excited or activated oxygen available to react with sputtering atom, and we believe this is the region where most of the reactive process occurs. This means that the metal-oxide film forms mainly in the high density plasma region, away from the target. The effect of this is to minimize the problems associated with "target poisoning". When the deposition rate is keep in a high growth rate" region of full oxide, it can be observed that this process was stable for long time running, without the need to adjust any of the deposition parameters and use any form of feed-back control system.

4. Conclusions

In this study a novel remote plasma sputtering technique for high rate deposition of Ta_2O_5 thin films was developed. It overcomes several of inherent limitations in conventional sputtering technology and realize the fully uniform erosion over the surface of the target. The deposition rate efficiency is almost four times larger than that of the conventional ion beam sputter method. By DC reactive sputtering, dense Ta_2O_5 thin films with a high deposition rate 0.53nm/s, near bulk material refractive index 2.152 at wavelength 510nm and low extinction coefficient 8.3×10^{-5} are achieved. The deposition rate and optical properties were studied with the process parameters change.

Acknowledgements

The authors are grateful to Prof. Mike and Dr. James from PlasmaQuestLtd UK for their technical assistance. This project was financially supported by the National Natural Science Foundation of China (No.50802080), and the Natural Science Foundation of Fujian Province of China (No.2010J01349).

References

[1] E. Atanassova, D. Spassov, A. Paskaleva, J. Optoelectron. Adv. Mater **9**(2), 315 (2007).
[2] C. C. Lee, D. J. Jan, Thin Solid Films, **483**(1-2), 130 (2005).
[3] J. C. Zhou, D. T. Luo, Y. Z. Li, L. Z. Liu, International Journal Of Modern Physics, **23**(27), 5275 (2009).
[4] Z. Todorova, N. Donkov, Z. Ristic, N. Bundaleski, S. Petrovic, M. Petkov, Plasma Process And Polymers, **3**(2), 174 (2006).
[5] W. J. Liu, X. J. Guo, C. H. Chien, Surface & Coatings Technology, **196**(1-2), 69 (2005).
[6] S. V. J. Chandra, P. S. Reddy, G. M. Rao, S. Uthanna, Optoelectron. Adv. Mater. - Rapid Comm. **1**(10), 496 (2007).
[7] E. Cetinorgu, B. Baloukas, O. Zabeida, J. E. Klemberg-Sapieha, L. Martinu, Applied Optics, **48**(23), 4536 (2009).
[8] C. Xu, C. Y. H. Qiang, Y. B. Zhu, J. D. Shao, Z. X. Fan, J. Optoelectron. Adv. Mater. **11**(8), 863 (2009).
[9] H. J. Qi HJ, X. D. Xiao, H. B. He, K. Yi, Z. X. Fan, Applied Optics, **48**(1), 127 (2009).
[10] W. M. Yang, Y. W. Liu, Q. Zhang , Y. X. Leng, H. F. Zhou, P. Yang, J. Y. Chen, N. Huang, Surface & Coatings Technology **201**, 8062 (2007).
[11] M. J. Thwaites, UK Patent GB 2 343 992 B: High Density Plasma (2001).
[12] S. J. Wakeham, M. J. Thwaites, B. W. Holton, C. Tsakonas, W. M. Cranton, D.C. Koutsogeorgis, R. Ranson, Thin Solid Films, **518**(4), 1355 (2009)
[13] M. Vopsaroiu, G. Vallejo Fernandez, M. J. Thwaites, J. Anguita, P. J. Grundy, Journal of Physics D: Applied Physics, **38**, 490 (2005).
[14] M. Nowak, Thin Solid Films, **254**, 200 (1995).
[15] S. Berg, T. Nyberg, Thin Solid Films, **476**, 215 (2005).

*Corresponding author: chennan@xmu.edu.cn