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Preparation and optical properties of SiO_x thin films using different reactive magnetron sputtering technology

Q. F. CHEN[a,b], Y. Y. LI[a, b], P. Y. YING[a,b], T. L. WANG[a,b], P. ZHANG[a,b *], J. B. WU[a,b], M. HUANG[a,b], Y. H. FANG[a,b], V. LEVCHENKO[a,b]
[a]*Zhejiang Provincial Key Laboratory for Cutting Tools, Taizhou University, Taizhou 318000, China*
[b]*School of Pharmaceutical and Materials Engineering, Taizhou 318000, China*

DC reactive magnetron sputtering (DC-RMS) and RF reactive magnetron sputtering (RF-RMS) have been employed to deposit SiO_x films under different O_2 content. Scanning electron microscopy (SEM) morphology indicates that the films as deposited possess a granular surface. According to the spectra, all films have good transmittance in the visible to near infrared region. Nevertheless, in the region close to ultraviolet, the films deposited by RF-RMS has better transmittance. The refractive index of the films prepared by DC-RMS and RF-RMS ranges from 1.43-1.47 and 1.46-1.47 at 800 nm respectively. The deposition rates of these two methods have similar trend. Anyway, SiO_x films with good optical performance could be prepared by these two methods under appropriate oxygen content during depositing.

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Keywords: SiO_x films, Reactive magnetron sputtering, Transmission property, Refractive index, Deposition rate

1. Introduction

Due to their excellent optical and physicochemical properties, SiO_2 films have been widely used in semiconductor, microelectronics, optics and other fields. In recent years, many methods such as ion beam sputtering (IBS) [1-3] and magnetron sputtering (MS) [4] have been developed to prepare high quality SiO_2 films for low-loss optical applications. IBS has been used in fabricating low optical elements such as high refractive mirrors due to the advantage of low absorption and scattering, stable optical properties and high-quality deposition [2-3]. However, low deposition rate and expensive equipment limit application of this method. Magnetron sputtering which is also adapt at preparing dense and smooth films [5] has been considered as a candidate to deposit high quality SiO_2 films.

Different magnetron sputtering such as radio frequency reactive magnetron sputtering [6], mid-mediate frequency reactive magnetron sputtering [7-9] and direct current reactive magnetron sputtering [10-12] have been employed to deposit SiO_2 films. In reactive sputtering, Ar/O_2 played an important role in the properties of the films as deposited. Therefore, it drew many attentions on the influence of O_2 on deposition rate, stoichiometry of Si/O and optical properties of SiO_x films [6, 7, 9, 11-13]. Stoichiometry of Si and O in SiO_x films deposited by RMS also has relation with sputtering angle [14]. However, there is few papers mentioned different reactive magnetron sputtering to prepare high quality SiO_2 films for optical application together. Here, DC reactive magnetron sputtering (DC-RMS) and RF reactive magnetron sputtering (RF-RMS) have been used to deposit SiO_x films. Scanning electron microscopy (SEM), spectrophotometer and spectral ellipsometer have been employed to test the surface morphology and optical properties of the films. The different optical properties of SiO_x films prepared by these two methods have been analyzed according to the sputter characteristics of such two reavtive magnetron sputtering technology.

2. Experimental

SiO_x films was deposited on monocrystalline silicon with a Miller index of (111) and fused silica in a vacuum coating system (DE500) with two sets of cathodes with DC power and one set of cathodes with RF power. High purity silicon (99.999%) was used as sputtering target. Argon (99.999%) and oxygen (99.999%) were used as working gas and reaction gas respectively. SiO_x films were synthetized under different O_2/Ar of 0.05, 0.10, 0.15 and 0.20. Before deposition, the substrates were cleaned by ultrasonic cleaning machine in acetone and alcohol for 20 minutes. Sputtering power, work pressure, the rotate speed of substrate and deposition time were 50W, 8 mTorr, 5 RPM and 3 hours respectively during depositing. All films were deposited under at room temperature. During depositing, the target was floating and the distance between the substrate and the target was about 18 cm. When the films were deposited by using DC-RMS, the stable voltage (current) of Si target is 392 V (0.128 A), 361 V (0.139 A), 333V (0.150 A), and 280V (0.179 A) with the O_2/Ar of 0.05, 0.10, 0.15 and 0.20

respectively.

After depositing, the surface morphology and cross section morphology of the samples were analyzed by using SEM (HITACHI, S-4800). The transmission spectra of the films were characterized by ultraviolet-visible-infrared spectrophotometer (Hitachi Limited, U-4100) with the wavelength ranges from 200 to 2000 nm. The refractive index and thickness of the films were characterized by spectral ellipsometer (M-2000 DI) range from 400 to 1200 nm. Then, the deposition rates were obtained by dividing the film thickness of the films to the deposition time.

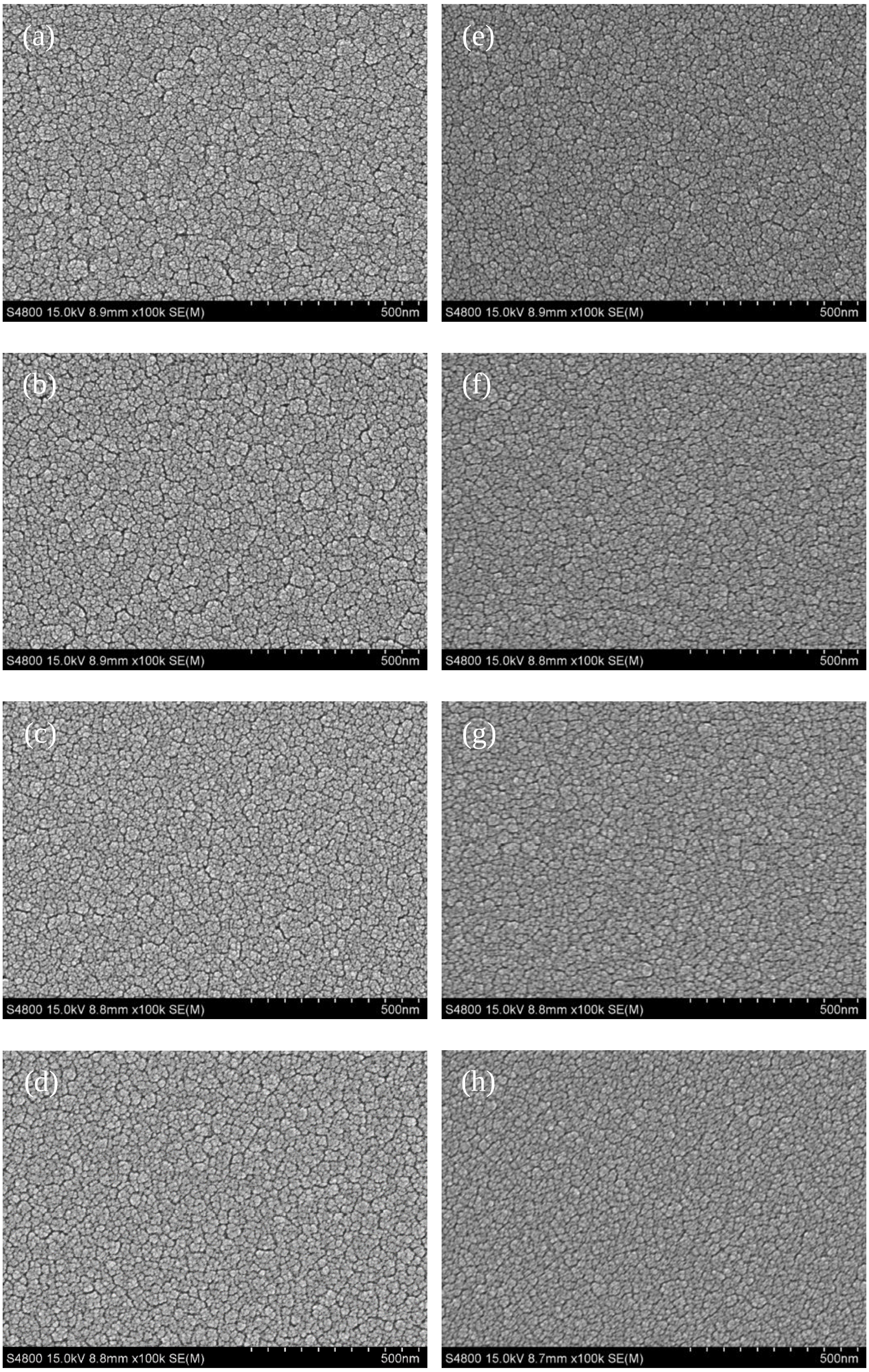


Fig. 1. Surface morphology of SiO_x films deposited by reactive magnetron sputtering: (a) DC, O_2/Ar = 0.05; (b) DC, O_2/Ar = 0.10; (c) DC, O_2/Ar = 0.15; (d) DC, O_2/Ar = 0.20; (e) RF, O_2/Ar = 0.05; (f) RF, O_2/Ar = 0.10; (g) RF, O_2/Ar = 0.15; (h) RF, O_2/Ar = 0.20

3. Results and discussion

Fig. 1 shows the surface morphology of SiO_x films deposited by DC-RMS and RF-RMS. As shown in the figure, (a), (b), (c) and (d) are the surface morphology images of SiO_x films prepared by DC-RMS with O_2/Ar values of 0.05, 0.10, 0.15 and 0.20 respectively. While, (e), (f), (g) and (h) are the surface morphology of the ones using RF-RMS with O_2/Ar values of 0.05, 0.10, 0.15 and 0.20. It can be seen that the films are compact with a granular surface. Meanwhile, the grains on the surfaces are uniform and very small with a size about 50 nanometers as shown in the images. The grains size gets reduced with the growth of O_2 content obviously according to the morphology. By comparison, the grains size of the films prepared by RF sputtering seems be smaller than the one deposited under DC sputtering. Moreover, it indicates that the films are more compact when using RF-RMS. Fig. 2 shows cross-section morphology of two SiO_x films samples. Among these two images, (a) is the morphology of the one prepared by DC sputtering under O_2/Ar of 0.1 and (b) is the morphology of the one using RF sputtering with the same ratio of O_2/Ar. From the figure, it can be seen that the films grow with columnar structure.

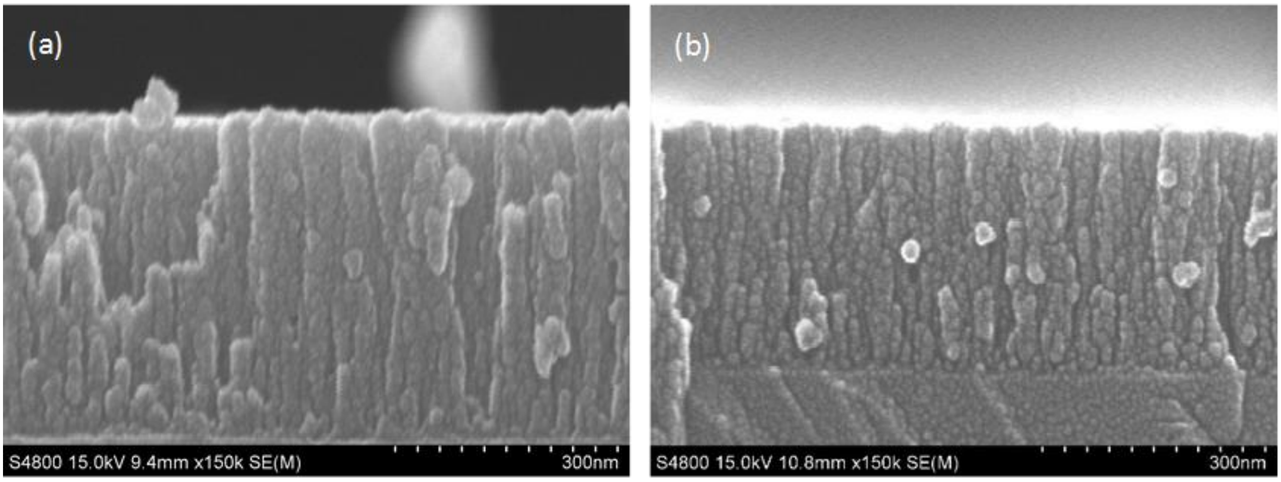
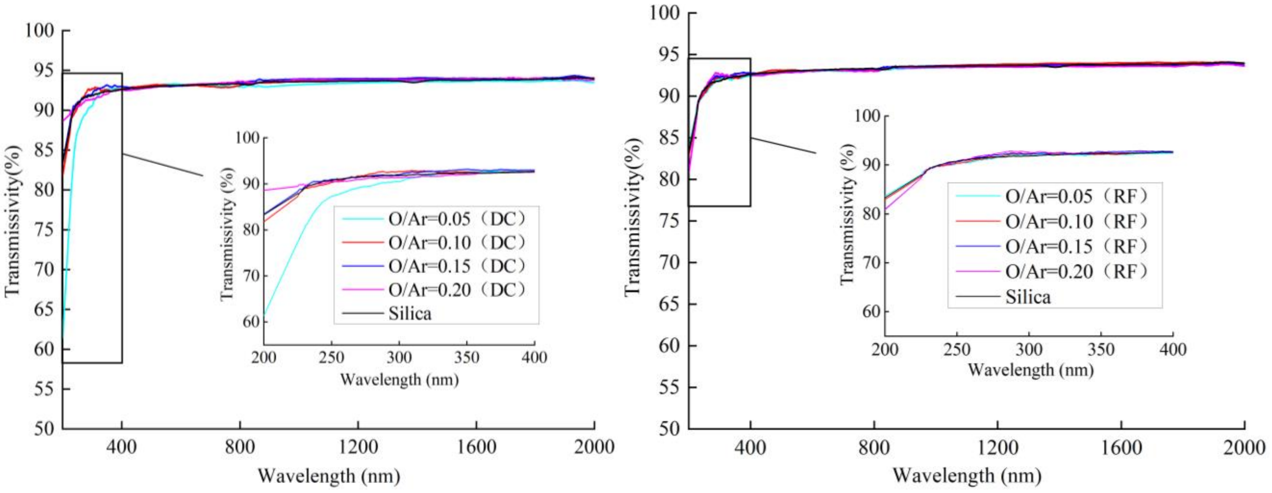


Fig. 2. Cross-section morphology of SiO_x films deposited by reactive magnetron sputtering.
(a) DC, O_2/Ar = 0.10; (b) RF, O_2/Ar = 0.10

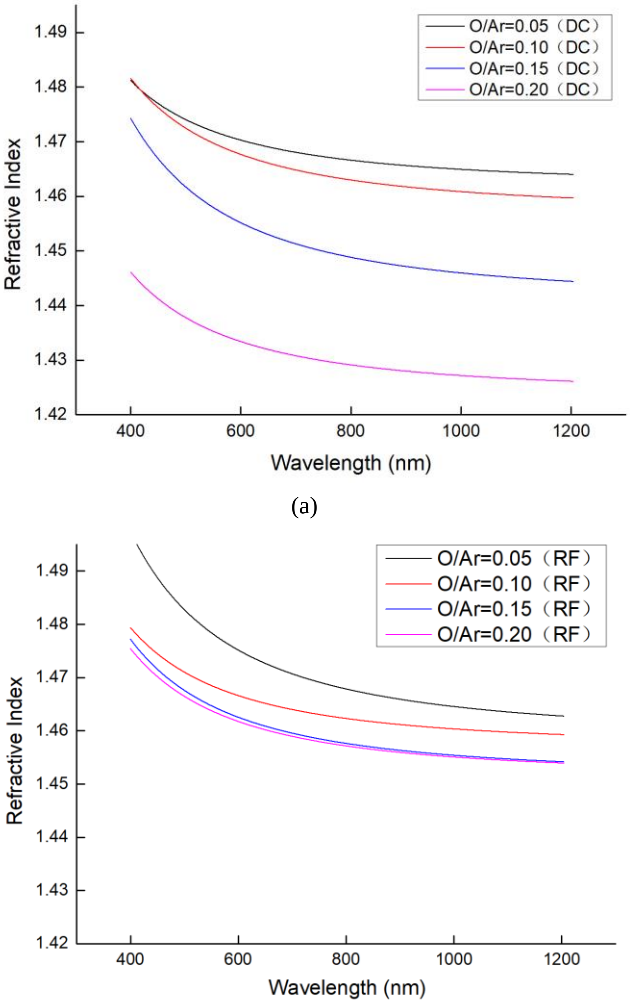


(a) (b)

Fig. 3. Transmissivity of SiO_x films prepared by DC (a) and RF (b) reactive magnetron sputtering (color online)

Transmission spectra ranging from 200 nm to 2000 nm wavelength of SiO_x films prepared by these two reactive magnetron sputtering are shown in Fig. 3. Transmissivity of all films is as high as fused silica substrate for the whole samples in the visible region as well as infrared region except for the one deposited by using DC-RMS with the O_2/Ar ratio of 0.05. Which means most of the SiO_x films prepared by these two methods under different oxygen content have good transmission property in the visible to near infrared region. However, in the region close to ultraviolet, the films have slightly poor transmission performance. As can be seen from the illustration in Fig. 3 (a), the transmittance becomes worse with the decrease of O_2 content for the films deposited by DC-RMS. Especially for the one got under the O_2/Ar of 0.05, there is absorption obviously in this region. The illustration in Fig. 3 (b) gives out the transmissivity of the films prepared by RF-RMS in the same region. It can be easily obtained that all of these films have close transmission performance and there is no evident decrease on transmissivity with the decrease of O_2 content. That may be caused by the higher sputtering rates of Si target when used DC-RMS. Therefore, with the same O_2 content, there will be more Si atom which couldn’t be fully oxidized in the film. Consequently, it could cause more chemical mismatch and defects in SiO_x films which can lead to more absorption and scattering when using DC-RMS with not enough O_2 [11].

Fig. 4 shows the refractive index of all SiO_x film samples with the wavelength ranging from 400 nm to 1200 nm. From the figure, it can be seen that the refractive index decreases firstly and eventually becomes flat with the increase of wavelength. The dependences of refractive index with wavelength is in accordance with Cauchy’s dispersion formula. Meanwhile, the value of refractive index decreases in a whole with the increase of O_2/Ar for SiO_x films both deposited by DC-RMS and RF-RMS. Generally, the refractive index of the film deposited by PVD will trend to the one of SiO_2 material with enough O_2 content. However, as shown in Fig. 4 (a), it can be obviously seen that the refractive index is abnormal for the film deposited under O_2/Ar of 0.20 using DC-RMS. It has a tendency to about 1.43 with the increase of wavelength. That is lower than the normal value of SiO_2 film that is ranging from 1.45 to 1.46. One explanation for this phenomenon is that excessive O_2 content can bring about more unreacted oxygen atoms or ions which will form lattice oxygen vacancies in SiO_x film when using DC-RMS to prepare SiO_x film [12]. Another explanation is that with more O_2 content, the energy of the sputtered particles would be lower due to higher probability to collide with O_2 which haven’t involved in sputtering and thicker oxide layer on the surface of Si target. All these could result in growth defects and reduce the film density. Therefore, the refractive index of the film is below than the normal value of SiO_2 material. Nevertheless, the refractive index of SiO_x film using RF-RMS has no similar phenomenon. As can be seen from Fig. 4 (b), with the increase of O_2 content, the value becomes smaller and trends to a value between 1.46 to 1.47 at wavelength of 800 nm.

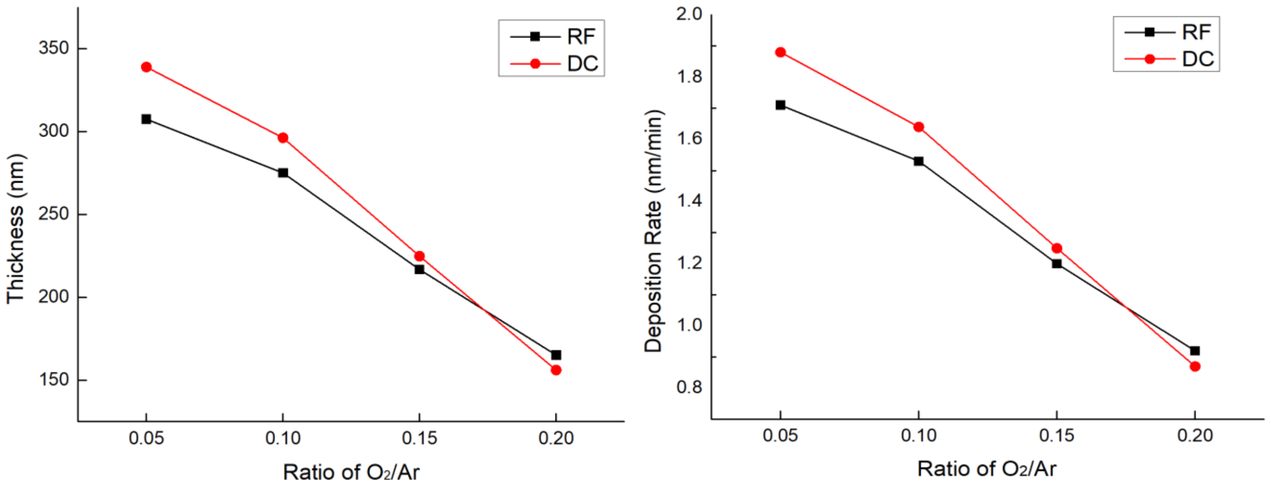


(a)

(b)

Fig. 4. Refractive index of SiO_x films prepared by DC-RMS (a) and RF-RMS (b) (color online)

The thickness and deposition rate of different films are shown in Table 1 and Fig. 5. As can be seen from the figure, the deposition rate decreases with the increase of the ratio of O_2/Ar for the films both deposited by DC-RMS and RF-RMS. From Fig. 5 (b), it can be obviously obtained that the deposited rate of DC-RMS is larger than the one of RF-RMS with a low O_2 content. Then, the gap between these two values becomes small with O_2 content increasing. Finally, the deposited rate of DC-RMS is lower than the one of RF-RMS when the ratio of O_2/Ar is 0.20. As is known to all, the reactive magnetron sputtering has three stages with the increasing of O_2 content. Silicon oxide would be formed on the surface of Si target due to silicon on the target surface could reactive with oxygen which is mixed in the working gas. When the O_2 content is very low, the sputtering rate would be larger than oxidized rate on the target surface. This stage is similar with the situation sputtering on Si target directly. With the O_2/Ar of 0.05, DC magnetron sputtering has a larger deposited rate than RF magnetron sputtering. That results in the deposited film may be not fully oxidized which lead to large absorption and scattering in visible near UV and UV region. When there is enough O_2 content, the surface will be completely oxidized. At this situation, DC-RMS has difficulty in sputtering the target surface which results in a low depositing rate. Thus, there are more unreacted oxygen atoms or ions to collide with sputtered particles. It would cause the film is not dense enough. As a result, the film deposited by DC-RMS with O_2/Ar of 0.20 possess lower refractive index than normal value of SiO_2 film. Unlike DC-RMS, RF-RMS could sputter the oxide layer on the target surface without any difficulty. In addition, there is a transition status between these two stages. In this condition, the target surface is only oxidized partly and the sputtering rate would be changing with the O_2 content. The oxide content on the target surface and in the films will both rise with the increase of O_2 content. That would cause a decrease of the sputtering rate for both RMS methods. Furthermore, based on the transmission spectrum and films thickness, the values of optical band gap were estimated to be 4.11 - 5.15 eV and 5.08 - 5.25 eV for the films obtained by DC-RMS and RF-RMS respectively. Which is a little lower than the one gotten by Mingdong Kong, et al [15]. This may be affected by the transmission of the substrate. It is best to use other substrates such as MgF_2 instead of fused silica to study the optical band gap of SiO_x films.



(a) (b)

Fig. 5. Thickness and deposition rate of SiO_x films prepared by DC-RMS (a) and RF-RMS (b) (color online)

Table 1. Thickness and deposition rate of SiO_x films deposited by RF-RMS and DC-RMS

Ration of O_2/Ar	Thickness		Deposition rate	
	RF-RMS (nm)	DC-RMS (nm)	RF-RMS (nm/min)	DC-RMS (nm/min)
0.05	307.424	338.900	1.71	1.88
0.10	275.020	296.245	1.53	1.64
0.15	216.739	224.811	1.20	1.25
0.20	165.190	156.093	0.92	0.87

4. Conclusion

SiO_x films have been deposited on Si and silica glass by DC-RMS and RF-RMS with different ratio of O_2/Ar respectively. The morphology has been tested by SEM and the optical properties have been studied using spectrophotometer and spectral ellipsometer. Whether the ones deposited by DC-RMS or by RF-RMS, the films possess granular surface according SEM morphology. The SiO_x films all have good transmission property in the visible to near infrared region. However, in the region close to ultraviolet, the ones prepared by RF-RMS have better transmission properties. According to analysis, this may be related to the higher deposition rate of DC magnetron sputtering with less oxygen content especially for the one deposited under the O_2/Ar of 0.05. The refractive index ranges from 1.43-1.47 for the films deposited by DC-RMS and 1.45-1.47 for the ones gotten by RF-RMS at 800 nm. The film with abnormal low refractive index was prepared by DC-RMS with the O_2/Ar of 0.20. This may have relation with the fact that the film structure prepared in this case is not dense enough. Moreover, the deposition rate had a same tendency and decreased with the increase of the ratio of

O_2/Ar for both DC-RMS and RF-RMS. With the O_2 content increasing, DC-RMS has a larger deposition rate because of its higher sputtering rate at a low O_2 content. Nevertheless, the deposited rate of RF-RMS becomes higher with enough O_2 content due to its advantages on sputtering oxides on the surface of Si target. In conclusion, DC-RMS and RF-RMS both could be used to prepare SiO_x films with good optical performance with appropriate oxygen content during depositing.

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*Corresponding author: zhangp03@126.com