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Studies of structural and optical properties of sputtered SiC thin films

ABSTRACT

The present study explored the deposition of amorphous silicon carbide (a-SiC) thin films on Si (100) and glass substrates using RF-magnetron sputtering. The sputtering power is changed from 100 to 250 W to study its influence on the characteristics of a-SiC thin films. Raman spectroscopy reveals the formation of a-SiC as well as carbon clusters. The film deposited at 100 W clearly shows the presence of both transverse optical (TO) and longitudinal optical (LO) phonon modes. The average roughness of the a-SiC films found to follow an increasing trend with increase in the sputtering power. The optical band gap of the a-SiC films measured by UV-Visible spectrophotometer was found to increase up to 2.45 eV with decrease in sputtering power. All a-SiC thin films were highly transparent. The Photoluminescence (PL) spectroscopy results were in agreement with the data observed by UV-Visible spectroscopy.
Keywords: *SiC thin films, Magnetron sputtering, Raman spectroscopy, PL spectroscopy*

1. INTRODUCTION

Silicon carbide (SiC) is a promising semiconductor material because of its excellent physical and chemical properties. Due to its fine tunable properties, SiC have attracted a great scientific and technological interest and have been used in many kinds of optoelectronic devices, such as solar cell windows layer, color sensors, and thin film light emitting and detecting devices [1-4]. In addition, amorphous SiC (a-SiC) thin films are chemically inert and are an excellent alternate passivation layer for silicon solar cells as well as MEMS application. SiC is hard material and at the moh scale it is just below from the diamond. Some other materials like TiAlBN have studied for their structural and hardness properties [5]. Thin films of a-SiC are usually synthesized by glow discharge method and consequently are hydrogenated a-SiC (a-SiC:H) [6]. To establish electrodes for electrical contacts, the passivity layer bears the high temperature firing process. However, during high temperature firing process, hydrogen in the a-SiC films evaporated. After the evaporation, many hydrogen molecules voids are generated. Therefore, the firing process brings degradation in the temperature stability of the films [7,8]. In this study, voids free a-SiC thin films have been deposited by RF magnetron sputtering on Si (100) substrate for solar cells and MEMS applications.

Several techniques are used for the synthesis of SiC thin films. Some of the common methods are chemical vapor deposition (CVD) [9,10], pulsed laser deposition (PLD) [11,12], molecular beam epitaxy [13,14] and reactive magnetron sputtering [15-17]. Among these techniques reactive magnetron sputtering is most attractive due to low substrate temperature deposition technique, good adhesion and less concentration of lattice defects [18].

In the present study we attempt to investigate the role of sputtering power on structural and optical properties of a-SiC thin films deposited at 400ºC. It has been observed that the sputtering power has a great effect on the properties of SiC thin films.

2. EXPERIMENTAL DETAILS

RF-magnetron sputtering technique has been used to grow a-SiC thin films on glass (Corning 1737) and Si (100) substrates. A commercially available silicon carbide target (SCI-Engineered Materials, USA) with 99.99 % purity was used for deposition of films. The diameter and thickness of

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the target were 50 mm and 5 mm, respectively. The substrate was cleaned in an ultrasonic bath using acetone and methanol. The Si substrate was passivated in 5 % HF solution to prevent the oxide formation on the Si surface. For the deposition of a-SiC films the sputtering pressure and substrate temperature were fixed at 10 mTorr and 400ºC, respectively. However the sputtering power was varied in the range from 100 W to 250 W. A turbo-based pumping system was used to achieve a base pressure of ~10^{-6} Torr. High-purity argon (99.99%) was used as sputtering gas. The distance between target and substrate was 6 cm and the deposition time for all the films was kept constant for 1 hour. Before each deposition the target was pre-sputtered for 5 min in order to ascertain the same state of the target. The sputtering parameters are shown in the table 1.

After deposition, the a-SiC thin films were characterized by X-ray diffractometer (Bruker D8 Advance), Raman spectrometer (Renishaw) and Atomic Force Microscopy (NT-MDT, model: NTEGRA) to study the structural properties. The optical properties of the a-SiC thin films have been studied by UV-Vis-NIR spectrophotometer (Cary 5000 Varian) and Photoluminescence spectrometer (Perkin Elmer LS 55). The thicknesses of the samples were examined by Surface Profilometer (Ambios Technology XP-200).

3. RESULTS AND DISCUSSION

Figure 1 shows the XRD pattern of a-SiC thin films deposited on Si (100) substrate at the substrate temperature of 400°C with varying sputtering power.

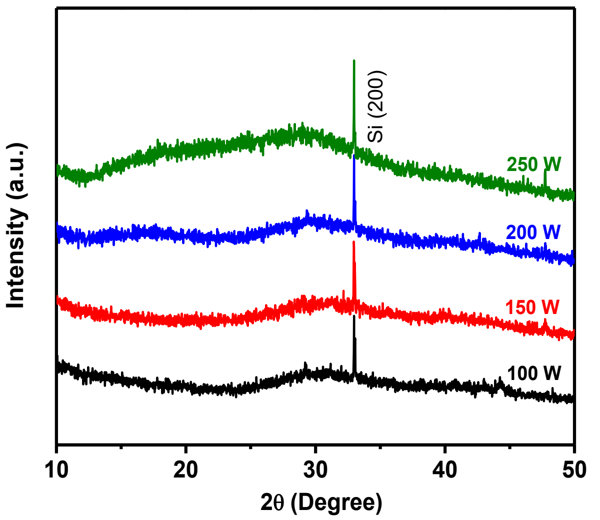

Figure 1. XRD pattern of a-SiC thin films deposited at various sputtering power

Slika 1. XRD uzorak a-SiC tankih filmova deponovanih pri različitoj snazi raspršivanja

Obviously no SiC diffraction peaks observed from the XRD patterns of the all samples, which show that the all samples are amorphous, because at the low substrate temperature there may not be sufficient energy to grow crystalline phase. The XRD peaks at angle (2θ ~ 33º) are corresponds to substrate peaks.

Figure 2 shows Raman spectra in the 100-1600 cm^{-1} region for the a-SiC thin films deposited on Si (100) substrate with varying sputtering power from 100 to 250 W. The Raman scattering measurements were performed at room temperature in the back-scattering configuration. Two broad bands are clearly observedin the regions of 600-1000 cm^{-1} and 1300-1600 cm^{-1} in the obtained Raman spectra. As the former band corresponds to the Raman band of a-SiC due to Si-C bond and the latter corresponding to that of amorphous carbon is due to C-C bonds [19].

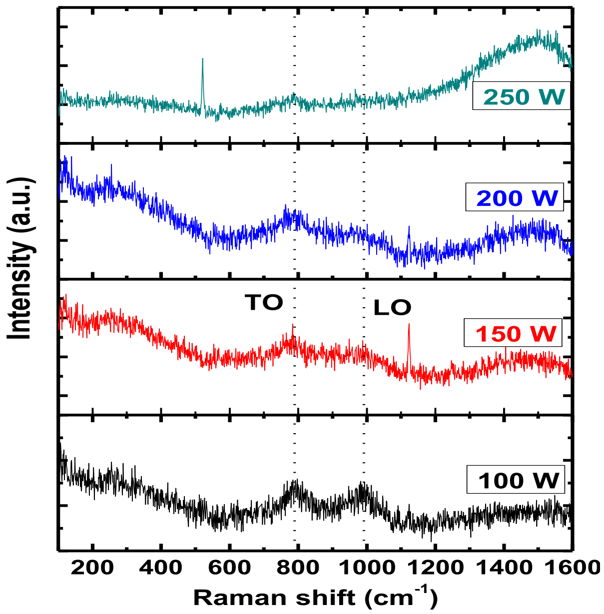

Figure 2. Raman spectra of a-SiC thin films deposited at various sputtering pressures (a) 5mTorr, (b) 10 mTorr, (c) 15 mTorr, (d) 20 mTorr and (e) 25 mTorr

Slika 2. Ramanski spektri a-SiC tankih filmova deponovanih pri različitim pritiscima raspršivanja (a) 5 mTorr, (b) 10 mTorr, (c) 15 mTorr, (d) 20 mTorr i (e) 25 mTorr

The film deposited at sputtering power of 100 W clearly shows two phonon modes, namely a transverse optical (TO) phonon mode (795 cm^{-1}) and a longitudinal optical (LO) phonon mode (970 cm^{-1}) at the Γ- point of the Brillouin zone centre. Presence of the C-clusters was arising due to the superimposition of E_{2g} stretching mode and A_{1g}

mode of small crystallites of graphite [20]. In this paper, the sputtering power has been optimized to synthesize a-SiC thin films with TO and LO modes and the intensities of both modes decreases with increasing of sputtering power. Raman peak in the 630-1000 cm^{-1} band was observed due to phonon damping. The reason for this phonon damping may be attributed to the short range ordering of SiC crystallites and the effect of surroundings having Si as well as C-clusters at higher sputtering power.

The AFM images of a-SiC thin-films deposited at varying sputtering power from 100 to 250 W on Si (100) substrates are shown in Figure 3. From the images we can observe that the thin-films grow in a way of columns or grains and the surface is very compact. The grains are uniformly distributed, and they are all in the shape of ellipse, which is consistent with Refs. [21]. It was observed that with increasing the sputtering power, the value of surface roughness increase. The average surface roughness of the films were observed from 8 nm to 11 nm with varying the sputtering power from 100 to 250 W, respectively as shown in Table 1.

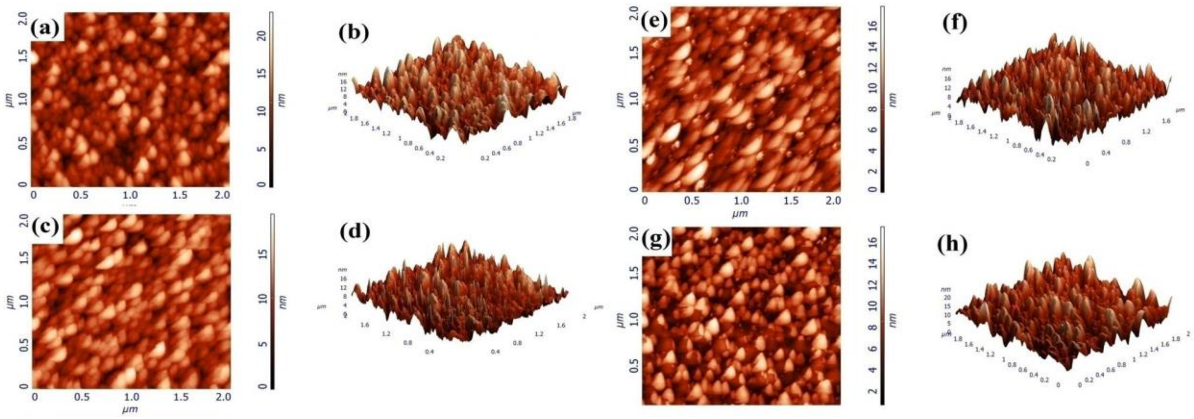

Figure 3. 2D and 3D AFM images of a-SiC thin films deposited at various sputteringpower (a)&(b) 100 W, (c)&(d) 150 W, (e)&(f) 200 W and (g)&(h) 250 W

Slika 3. 2D i 3D AFM slike a-SiC tankih filmova deponovanih pri različitim snagama raspršivanja (a)&(b) 100 V, (c)&(d) 150 V, (e)&(f) 200 V i (g) i (h) 250 V

These smaller values of surface roughness indicate that the films were very smooth and have good adherence to the substrate. The surface roughness increases due to the increase in grain size with increasing the power from 100 to 250 W. In order to understand the effect of sputtering power it is necessary to understand the basic working principle of the rf magnetron sputtering. In sputtering process, Argon gas ionized and the argon ions hit the target due to positive charge on the ion and negative charge on the target. In this process the target atoms ejected and deposited on the substrate. If the substrate temperature is low then the sputtered atoms have not much time to move on the substrate surface. We have deposited a-SiC at the temperature of 400 °C which provide sufficient energy to the sputtered atoms to move on the surface which leads to grow in the column-shaped structure. When sputtering power increased to 150 W both the kinetic energy and deposition rate increased significantly. So the size and the shape has been significantly changed which leads to increase in the thickness of the films [22]. Further increase in the sputtering power up to 250 W, the sputtered atoms have the more kinetic energy which leads to the more increase in surface roughness. We have chosen the sputtering pressure 10 mTorr for all the depositions because we have optimized it in previous study [23].

Table 1.Properties of SiC thin films with variation of sputtering power

Tabela 1. Osobine tankih filmova SiC sa varijacijama snage raspršivanja

S. No.	Sputtering power (W)	Band gap (eV)	Refractive index	Thickness (nm)	Average Transmittance (%)	Average roughness (AFM) (nm)
1.	250	2.21	1.54	1088	84	11
2.	200	2.33	1.65	992	87	10
3.	150	2.40	1.69	967	89	9
4.	100	2.45	1.98	962	89	8

The spectral variations of transmission for the a-SiC films deposited onto glass substrate at various deposition pressure were measured over the wavelength range 200–1100 nm and are shown in Figure 4.

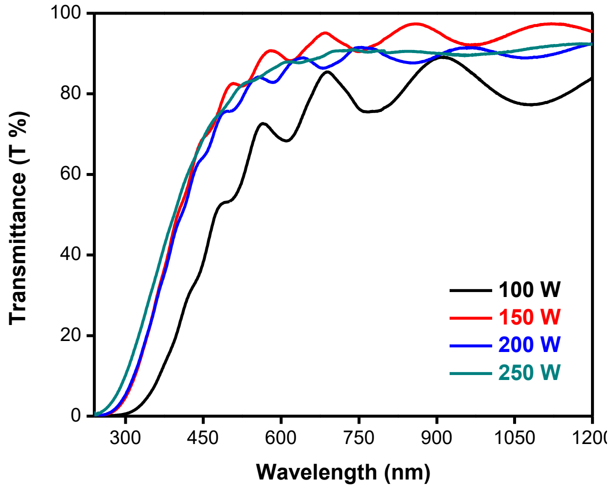

Figure 4. Optical transmittance spectra of a-SiC thin films deposited at various sputtering pressure

Slika 4. Spektri optičke propusnosti a-SiC tankih filmova deponovanih pri različitim pritiscima raspršivanja

The films show very good transmittance in the visible range. It can be clearly seen that transmittance decreases with decreasing of sputtering pressure. As described above, with increase in sputtering pressure, the surface of the films becomes rough and hence results in more scattered light and less transmittance. The film deposited at low sputtering power shows a high transmittance because of minimal scattering light loss caused by the rough surface [24]. The scattering light loss increases with increasing of sputtering power which leads to decrease in transmittance.

Refractive index (n) was estimated from the transmission spectrum by using envelope method and following expression was used to calculate the refractive index.

$$n = [N + (N^2 - n_0^2 n_1^2)^{1/2}]^{1/2}$$

where

$$N = \left[\frac{n_0^2 + n_1^2}{2}\right] + 2\, n_0 n_1 (T_{max} - T_{min})/T_{max}T_{min}$$

n_o is the refractive index of air, n_1 is the refractive index of substrate, T_{max} and T_{min} are maximum and minimum transmittance values at the same wavelength. The calculated refractive indices for the film deposited at 400 °C are shown in Table 1. The refractive indices were found to be decrease with decreasing of sputtering pressure up to a limit and increase further decreasing of sputtering pressure.

The absorption spectra of a-SiC thin film have been recorded as a function of the photon energy (hv) in the wavelength range of 200 to 1100 nm. In order to calculate the indirect optical band gap (E_g) of a-SiC thin film deposited at different sputtering pressure we have used the Tauc relation [25].

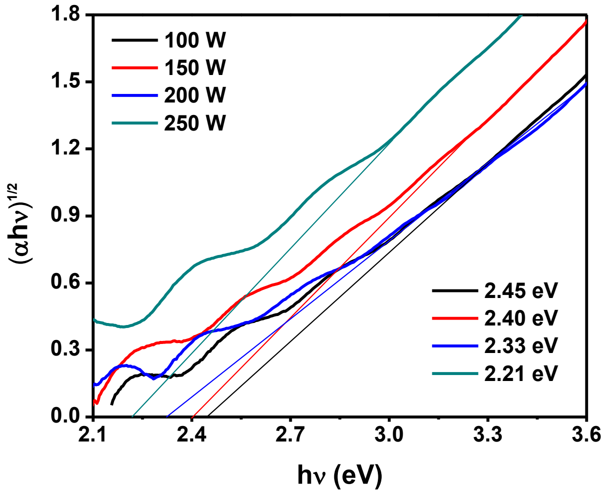

Figure 5. $(\alpha h\nu)^{1/2}$ versus $(h\nu)$ plot of a-SiC thin films deposited on glass substrate at various sputtering pressure

Slika 5. Grafikon (ahn)1/2 u odnosu na (hn) tankih filmova a-SiC nanesenih na staklenu podlogu pri različitim pritiscima raspršivanja

The absorption spectra of a-SiC thin film have been recorded as a function of the photon energy (hv) in the wavelength range of 200 to 1100 nm. In order to calculate the indirect optical band gap (E_g) of a-SiC thin film deposited at different sputtering pressure we have used the Tauc relation [25].

The band gaps of the a-SiC thin films deposited at sputtering power 100, 150. 200 and 250 W was calculated 2.45 eV, 2.40 eV, 2.33 eV and 2.21 eV, respectively as shown in Figure 5 and Table 1**.** The band gap increase with decrease in the sputtering power up to 250 W could be due to increase in grain size as explained above in the AFM results (shown in Figure 3).

Figure 6 depicts the room temperature Photoluminescence (PL) spectra of the a-SiC thin films deposited on Si (100) at different sputtering pressure. The excitation wavelength for all a-SiC thin films was 450 nm. PL spectrum of a-SiC thin films deposited at 250 W shows a broad and intense peak at ~ 532 nm near the band gap

emission. This emission was attributed to the band edge emission from the a-SiC [26]. The band gap of SiC has been calculated using thin emission and found to be consistent with the values obtained from optical absorption. As the sputtering power decrease to 200 W, two PL peaks were observed. The one is located at 526 nm, and the other is located at 604 nm. Further decreasing of sputtering power to 150 W both PL peaks red shift to 528 nm and 609 nm respectively and the intensity of 609 nm peaks is low. It can be seen clearly that the PL peaks have been found to red shift by increasing the sputtering power from 100 W to 250 W. These PL spectra are similar to those observed SiC thin films and $SiO_2/Si/SiO_2$ structure [27]. The PL results are in accordance with the UV-visible spectroscopy results.

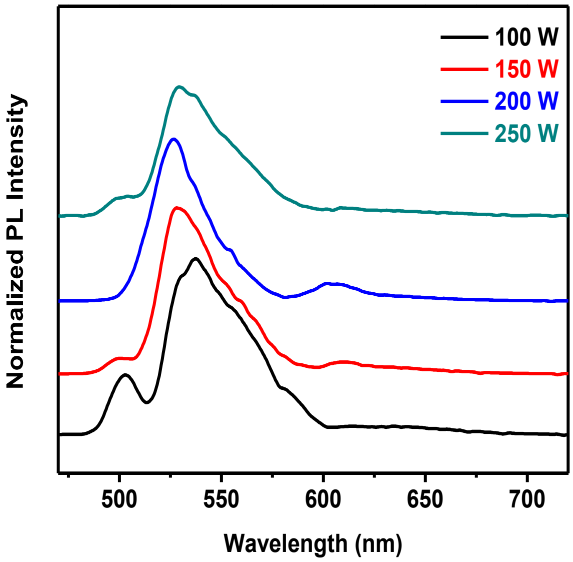

Figure 6. Photoluminescence spectra of a-SiC films deposited at various sputtering pressure

Slika 6. Spektri fotoluminiscencije a-SiC filmova deponovanih pri različitim pritiscima raspršivanja

Figure 6 depicts the room temperature Photoluminescence (PL) spectra of the a-SiC thin films deposited on Si (100) at different sputtering pressure. The excitation wavelength for all a-SiC thin films was 450 nm. PL spectrum of a-SiC thin films deposited at 250 W shows a broad and intense peak at ~ 532 nm near the band gap emission. This emission was attributed to the band edge emission from the a-SiC [26]. The band gap of SiC has been calculated using thin emission and found to be consistent with the values obtained from optical absorption. As the sputtering power decrease to 200 W, two PL peaks were observed. The one is located at 526 nm, and the other is located at 604 nm. Further decreasing of sputtering power to 150 W both PL peaks red shift to 528 nm and 609 nm respectively and the intensity of 609 nm peaks is low. It can be seen clearly that the PL peaks have been found to red shift by increasing the sputtering power from 100 W to 250 W. These PL spectra are similar to those observed SiC thin films and $SiO_2/Si/SiO_2$ structure [27]. The PL results are in accordance with the UV-visible spectroscopy results.

4. CONCLUSION

In summary, a-SiC thin films were successfully grown on two substrates namely Si (100) and glass. The effect of sputtering power on structural and optical properties of a-SiC thin films were studied. No SiC peaks found in XRD pattern which show the amorphous nature of all samples deposited at different sputtering power ranging from 100 to 250 W. The amorphous nature of SiC has been confirmed by Raman spectrometer and both TO and LO modes were clearly present in the Raman spectra with very less carbon concentration in a-SiC films deposited at 100 W. The optical band gap of the a-SiC films was found to increase up to 2.45 eV with decrease in sputtering power from 250 to 100 W. The transmittance of the a-SiC films was found to decrease with increasing the sputtering power. Further it was observed that the band gap decreases with increase in sputtering power as confirmed by UV-visible and PL spectra. Thus, the sputtering parameters were found to have a great influence on the structural and optical properties of a-SiC thin films.

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IZVOD

STUDIJE STRUKTURNIH I OPTIČKIH OSOBINA RASPRŠENIH SiC TANKIH FILMOVA

Ova studija je istraživala taloženje tankih filmova amorfnog silicijum karbida (a-SiC) na Si (100) i staklene podloge korišćenjem RF-magnetronskog raspršivanja. Snaga raspršivanja je promenjena sa 100 na 250V da bi se proučavao njen uticaj na karakteristike a-SiC tankih filmova. Ramanova spektroskopija otkriva formiranje a-SiC kao i klastera ugljenika. Film deponovan na 100 V jasno pokazuje prisustvo i poprečnih optičkih (TO) i uzdužnih optičkih (LO) fononskih modova. Utvrđeno je da prosečna hrapavost a-SiC filmova prati rastući trend sa povećanjem snage raspršivanja. Utvrđeno je da se optički razmak a-SiC filmova meren UV-vidljivim spektrofotometrom povećava do 2,45 eV sa smanjenjem snage raspršivanja. Svi a-SiC tanki filmovi bili su veoma providni. Rezultati fotoluminiscencije (PL) spektroskopije bili su u saglasnosti sa podacima uočenim UV-vidljivom spektroskopijom.

Ključne reči: SiC tanki filmovi, Magnetronsko raspršivanje, Ramanova spektroskopija, PL spektroskopija

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