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An investigation on the effect of high partial pressure of hydrogen on the nanocrystalline structure of silicon carbide thin films prepared by radio-frequency magnetron sputtering



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HIGHLIGHTS

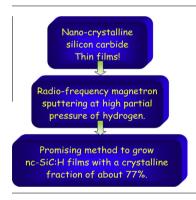
- nc-SiH:H are deposited by radiofrequency magnetron technique (RFMS).
- RFMS is a simple and promising method to grow nc-SiC:H films.
- H₂ dilution in the gas mixture strongly affects the growth of nc-SiC:H films.
- The nanocrystalline silicon is present in all series with a variable crystallite size.

ARTICLE INFO

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G R A P H I C A L A B S T R A C T



ABSTRACT

The aim of the study reported in this paper is to investigate the role of the high partial pressure of hydrogen introduced during the growth of nanocrystalline silicon carbide thin films (nc-SiC:H). For this purpose, we report the preparation as well as spectroscopic studies of four series of nc-SiC:H obtained by radio-frequency magnetron sputtering at high partial pressure of hydrogen by varying the percentage of H₂ in the gas mixture from 70% to 100% at common substrate temperature ($T_S = 500$ °C). The effects of the dilution on the structural changes and the chemical bonding of the different series have been studied using Fourier transform infrared and Raman spectroscopy. For this range of hydrogen dilution, two groups of films were obtained. The first group is characterized by the dominance of the crystalline phase and the second by a dominance of the amorphous phase. This result confirms the multiphase structure of the grown nc-SiC:H thin films by the coexistence of the Si—C network, carbon-like and silicon-like clusters. Furthermore, infrared results show that the Si—C bond is the dominant absorption peak and the carbon atom is preferentially bonded to silicon. The maximum value obtained of the crystalline fraction is about 77%, which is relatively important compared to other results obtained by other techniques. In addition, the concentration of CH_n bonds was found to be lower than that of SiH_n for all series.

* Corresponding author. Fax: +1 780 492 8231. *E-mail address: rekik@ualberta.ca* (N. Rekik). Raman measurements revealed that the crystallization occurs in all series even at 100% H₂ dilution suggesting that high partial pressure of hydrogen favors the formation of silicon nanocrystallites (nc-Si). The absence of both the longitudinal acoustic band and the transverse optical band indicate that the crystalline phase is dominant.

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Introduction

During the last decade, silicon carbide (SiC) has been extensively studied due to its wide applications [1–11]. Its unique physical and optical properties such as a wide band gap, excellent thermal conductivity, high breakdown electric field, and high saturated electron drift velocity make it a suitable semiconductor material for operating at high temperature, high frequency, high power, and chemically hostile environment. Because of the expected improvement in the performance of the devices upon crystallization of the SiC layer, several attempts were made so far reporting processing temperatures exceeding 500 °C [4] to prevent the generation of defects at the interface. The lowering of the deposition temperature of crystallized silicon carbide is, therefore, of paramount importance and we succeeded recently [5] in the achievement of nc-SiC:H layers with $T_{\rm S}$ as low as 500 °C (200 °C $\leq T_S \leq$ 500 °C), even though the crystalline quality and the crystallization degree of these layers are well, and both improved when T_S is increased up to 500 °C. In that study, we have demonstrated that a low substrate temperature ($T_S \leq 500 \,^{\circ}\text{C}$) deposition of nc-SiC:H thin films by radiofrequency magnetron sputtering (RFMS) technique was possible. In the process of synthesis of nc-SiC films using a precursor gas heavily diluted with hydrogen (Ar + H₂), we have found out that amorphous and crystalline phases are assumed to be deposited simultaneously.

In this work, we focused on the study of the affect of different hydrogen dilutions on the deposition of nanocrystalline SiC films. Indeed, the influence of hydrogen dilution on the growth of nanocrystalline SiC using RFMS will be investigated in detail. More specifically, the main goal of our study is to deposit nanocrystalline silicon carbide films (nc-SiC:H) with well-controlled properties and to understand the influence of hydrogen dilution on the growth of nanocrystalline SiC, nanocrystalline silicon size and the formation of graphitic domains in our films elaborated by the RFMS technique. The influence of the hydrogen dilution on the growth of nanocrystals will be studied using Fourier transformation infrared (FTIR) spectroscopy and Raman spectroscopy.

The structure of the letter is as follows: Section 'Experimental details' contains an overview of the experimental details of the preparation as well as the techniques used for the characterization of the samples. In Section 'Results and discussion', we present and discuss the experimental results, and conclusions are made in Section 'Conclusions'.

Experimental details

Four series of nanocrystalline hydrogenated silicon carbide labeled I to IV were deposited by radiofrequency (RF) plasmaassisted magnetron sputtering RFMS using a single crystal silicon target in an (Argon + X% H₂) gas mixture. The H₂ percentage in the gas phase varies from 70% to 100% H₂ at a common high RF power (500 W), high rates (11–15 Å/s) and a substrate temperature T_S fixed at 500 °C. The plasma is initiated between the cathode and the anode at a pressure of 50 mTorr by the application of a high "RF" voltage.

The IR absorption measurements were carried out between 400 and 4000 cm^{-1} by means of a Fourier-Transform Infrared

Spectrometer (Nicolet 750-II spectrometer) (FTIR) with a resolution of 2 cm^{-1} and averaged over 500 scans. IR absorption measurements were used to determine chemical bonding of hydrogen. The concentrations of different of hydrogen-bonded groups were obtained from the integrated intensity and by using the proportionality constants previously reported [12,13].

Raman spectra were recorded with a DILOR (Z24) Raman spectrophotometer equipped with a triple monochromator and watercooled photomultiplier and a photon counter system apparatus in the range 200–1200 cm⁻¹. The spectrometer has backscattering geometry for detection of Raman spectrum with the resolution of 1 cm⁻¹. The excitation source was a 514.5 nm line of an Argon ion laser. The power of the Raman laser was kept less than 50 mW to avoid laser induced crystallization on the films. The spot-size was about 80 μ m diameter.

Results and discussion

The chemical bonding of the samples are investigated by FTIR spectroscopy. Fig. 1 shows the FTIR absorption spectra of all series in the spectral region of 400–3500 cm⁻¹ deposited at different hydrogen dilution ratios % H_2 in the as deposited state. In order to compare the content of each bond among the films, FTIR spectra were normalized by the film thicknesses.

As expected [14], the spectra are characterized by three regions: $500-1200 \text{ cm}^{-1}$, $2000-2300 \text{ cm}^{-1}$ and $2800-3000 \text{ cm}^{-1}$. The band in the range $500-1100 \text{ cm}^{-1}$ is attributed to the superposition of four different vibration modes namely Si—C stretching around $760-795 \text{ cm}^{-1}$, Si—H bending near $800-900 \text{ cm}^{-1}$, and C—H_n rock-ing/wagging vibrating modes at about 1000 cm^{-1} , respectively. We have proceeded to perform the deconvolution of the predominant peak attributed to the Si—C into Gaussian and Lorentzian components after subtraction of the contributions of the SiH and C—H vibrational modes [15] in order to estimate the infrared crystalline volume fraction F_C . As we can see from Fig. 2, for all series, the Si—C band is the dominant absorption peak and represents about 35% of the total band area showing that the carbon atom is preferentially

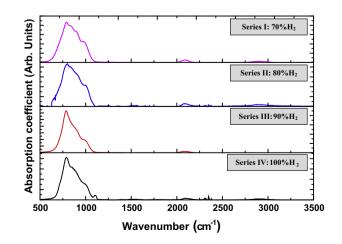


Fig. 1. FTIR absorption spectra of samples deposited at different hydrogen dilution percentage.

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