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Low refraction properties of F-doped SiOC:H thin films prepared by PECVD

S.G. Yoon [a,*], S.M. Kang [a], W.S. Jung [a], H. Kim [a], S.-W. Kim [b], D.H. Yoon [a]

[a] Department of Advanced Materials Engineering, Sungkyunkwan University, Suwon 440-746, Republic of Korea
[b] School of Advanced Materials and System Engineering, Kumoh National Institute of Technology, Gyeongbuk 730-701, Republic of Korea

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Abstract

Low refractive index materials which F-doped SiOC:H films were deposited on Si wafer and glass substrate by low temperature plasma enhanced chemical vapor deposition (PECVD) method as a function of rf powers, substrate temperatures, gas flow ratios (SiH_4, CF_4 and N_2O). The refractive index of the F-doped SiOC:H film continuously decreased with increasing deposition temperature and rf power. As the N_2O gas flow rate decreases, the refractive index of the deposited films decreased down to 1.378, reaching a minimum value at an rf power of 180 W and 100 °C without flowing N_2O gas. The fluorine content of F-doped SiOC:H film increased from 1.9 at.% to 2.4 at.% as the rf power was increased from 60 W to 180 W, which is consistent with the decreasing trend of refractive index. The rms (root-mean-square) surface roughness significantly decreased to 0.6 nm with the optimized process condition without flowing N_2O gas.

Keywords: F-doped SiOC:H; Refractive index; Chemical vapor deposition

1. Introduction

F-doped SiOC:H film having low refractive index and dielectric constant has attracted a lot of interests for the applications of antireflection coatings and ultra-large scale integrated (ULSI) devices due to its excellent transparency, and chemical and thermal stability with standard microelectronic fabrication processes [1]. Low refractive index materials can decrease the number of the optical coating layer and increase the emission efficiency by matching the refractive indices between ambient air and emitting layer of the optical device. Antireflection coatings for the visible and near infrared regions play an important role in the development of flat panel displays (FPDs) and solar cell systems, in which efficient optical properties, such as low reflection loss, wider bandwidth, high transmission efficiency, and high durability against adverse terrestrial and space conditions are required [2,3]. Therefore, the substrate has to have a refractive index which is sufficiently higher than that of the available thin film material for it to be possible to design high performance antireflection coatings consisting entirely, or almost entirely, of layers having lower refractive indices than that of the substrate. Color displays based on organic light emitting devices (OLED) deposited on flexible substrates are being developed as a promising alternative to liquid crystal displays (LCD). The substrate for flexible OLED is a multilayer composite structure having a couple of functional coatings on a polymer-based substrate. For the fabrication of light emitting diodes (LEDs) and organic LEDs, semiconducting materials, such as Ge and Si are commonly used as substrates. Typically, germanium having a refractive index of 3.9 gives a reflection loss of around 36% and silicon having a refractive index of 3.5 gives a reflection loss of 31% [4,5]. Among many possible deposition techniques in obtaining refractive index materials, plasma enhanced chemical vapor deposition (PECVD) is considered to be an outstanding method because it provides an easy controllability of the film stoichiometry and refractive index, and excellent surface roughness [6–10].

In this study, the deposition characteristics of F-doped SiOC:H films grown by PECVD were investigated as a function of process parameters, such as flow ratio of mixing gases, rf power, and the adding amount of N_2O gas. In addition, the relationship between the fluorine contents in the F-doped SiOC:H thin film and the refractive indices in the visible range (632.8 nm) was discussed.

2. Experimental procedure

F:SiOC:H films were deposited by PECVD technique [11] using appropriate gaseous mixtures of silane (H_2 90% dilution,

* Corresponding author. Tel.: +82 31 290 7388; fax: +82 31 290 7371.
E-mail address: sgyoon@skku.edu (S.G. Yoon).

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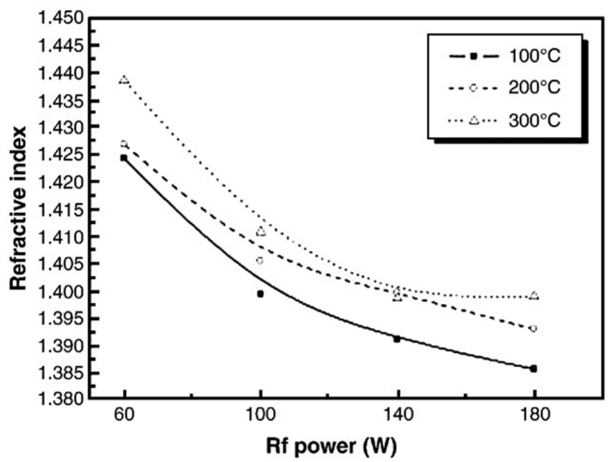

Fig. 1. The refractive indices of F-doped SiOC:H films as a function of rf power and deposition temperature.

SiH_4), nitrous oxide (99.999% N_2O) and tetrafluoromethane (99.999% CF_4). The reactor is a parallel planar discharge type having a rectangular rf (13.56 MHz) electrode (lower). The upper electrode supporting the substrate is connected to a 13.56 MHz rf bias power supply. The substrate is placed on a tray with the surface to be coated facing downward, so that the possible depositions of dust particles and flakes can be minimized. Si and glass were used as substrates and, prior to the F:SiOC:H film deposition, a short in-situ pre-cleaning and plasma pre-treatment of the substrate was performed in order to improve the adhesion of the film to the substrate using H_2 discharge at an rf power of 60 W. Because the refractive index of thin film strongly depends on the deposition conditions, the influence of the main deposition parameters, such as mixing gas flow ratio, rf power, and $N_2O/(SiH_4+CF_4)$ ratio, on the qualities of F-doped SiOC:H thin films was systematically studied. The pressure for all the deposition conditions was held at a constant value of 1.3×10^2 Pa throughout the experiment. After the deposition of F-doped SiOC:H films under various process conditions, the refractive index was measured using a prism

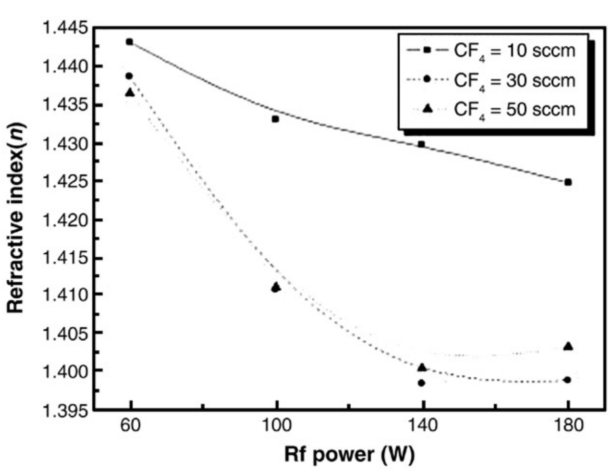

Fig. 2. The refractive indices of F-doped SiOC:H films as a function of rf power and CF_4 gas flow.

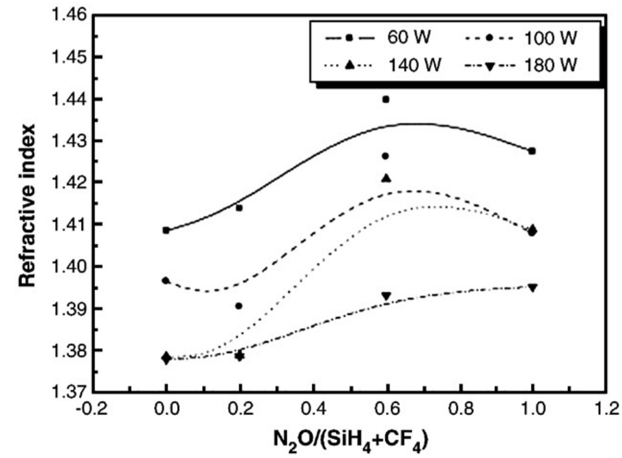

Fig. 3. The variations of the refractive indices of F-doped SiOC:H films as a function of $K=N_2O/(SiH_4+CF_4)$ and rf power at a constant deposition temperature.

coupler operating at a wavelength of 632.8 nm. X-ray photoelectron spectroscopy (XPS) and atomic force microscopy (AFM) were used to characterize the chemical binding state and the surface morphology of the films, respectively.

3. Results and discussion

F-doped SiOC:H films were deposited by low temperature PECVD using an SiH_4, CF_4, and N_2O gas mixture on glass substrates pre-treated with H_2 plasma. Fig. 1 shows the variation of the refractive indices of F-doped SiOC:H films as a function of rf power and deposition temperature. With the increase of deposition temperature from 100 °C to 300 °C, the refractive indices of all the samples increased. The lowest refractive index of F-doped SiOC:H film (1.385) was obtained at the deposition temperature of 100 °C and the rf power of 180 W. The variations of refractive indices with different rf power levels exhibited a similar trend with different deposition temperatures. For all of

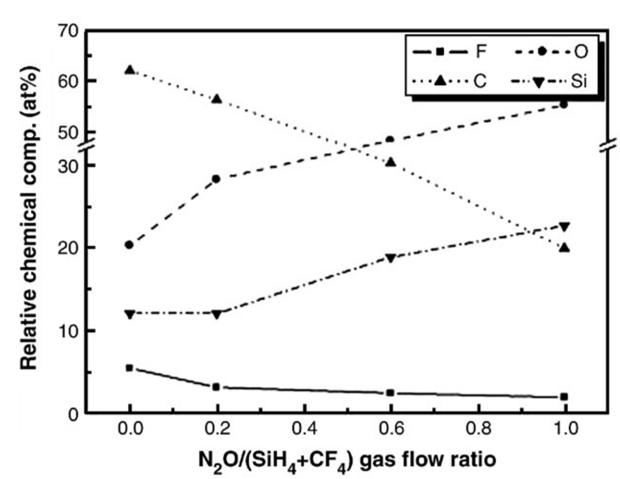

Fig. 4. The relative chemical composition of F-doped SiOC:H films from the XPS spectra as a function of $K=N_2O/(SiH_4+CF_4)$ gas flow ratio at a constant deposition temperature (100 °C) and rf power (180 W).