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PHOTO- AND ELECTRO-LUMINESCENCE FROM HYDROGENATED AMORPHOUS SILICON CARBIDE FILMS PREPARED BY USING ORGANIC CARBON SOURCE*

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Hydrogenated amorphous silicon carbide (a-SiC:H) films were grown by using an organic source, xylene (C_8H_{10}), instead of methane (CH_4) in a conventional plasma enhanced chemical vapor deposition system. The optical band gap of these samples was increased gradually by changing the gas ratio of C_8H_{10} to SiH_4. The film with high optical band gap was soft and polymer-like and intense photoluminescence were obtained. Room temperature electro-luminescence was also achieved with peak energy at 2.05 eV (600 nm) for the a-SiC:H film with optical band gap of 3.2 eV.

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I. INTRODUCTION

Hydrogenated amorphous silicon carbide (a-SiC:H) films have attracted much attention since they can be utilized in many kinds of opto-electronic devices, such as light emission diode and a-Si:H-based solar cells.[1] Generally, a-SiC:H films were prepared by using methane (CH_4) gas together with silane (SiH_4). However, it was reported that the optical band gap increases firstly to about 3.0 eV by increasing the carbon content in the film and then decreases due to the formation of graphite-like structures by further increasing the carbon content. Moreover, the intensity of emission light becomes weaker and weaker with increasing optical band gap because of the increase of the defect-state density in the films.[2,3]

In order to improve the optical and photoelectric properties of a-SiC:H alloys, some novel carbon sources have been used.[4,5] Recently, it was reported that an organic source, xylene (C_8H_{10}), could be used to deposit a-SiC:H alloys and blue light emission was observed at room temperature from these films.[6,7] However, it seems that the photoluminescence (PL) peak was not changed although the optical band gap of a-SiC:H films varied in a quite wide range and electroluminescence (EL) was still not realized. In this paper, we report the fabrication of a series of a-$Si_{1-x}C_x$:H ($0 < x \leq 1$) films by using organic source, xylene, in conventional plasma-enhanced chemical vapor deposition (PECVD) system. The optical properties were studied and the systematic measurements on PL at room temperature were performed. An intense PL could be observed and the PL peak energy was shifted with the carbon content in the films. Moreover, room temperature EL has been observed for our samples, which can be seen by naked eyes in the dark.

II. EXPERIMENTAL

Xylene and silane were used as the source gases to prepare a series of a-SiC:H films in diode-type PECVD system with different gas flow rate ratios. Liquid xylene source was kept at a constant temperature of 100 ℃ and the xylene vapor was carried into the reaction chamber by argon gas. The substrate temperature and the rf power is 200 ℃ and 15 W, respectively. It is noted that the rf density used in the present study is higher than that used in the previous work.[6] During the deposition, the SiH_4 flow rate is kept at 5 sccm and the flow rate of C_8H_{10}+Ar is changed. The total reaction pressure is about 150×133.322 mPa. The films were deposited on Corning 7059 glasses and fused quartz plates for optical measurements and on crystalline Si wafers for Fourier transform infrared (FT-IR) and photoluminescence measurements.

III. RESULTS AND DISCUSSION

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The Tauc gap, which is usually used to determine the optical band gap of amorphous materials, was deduced by using the following formula:[8]

$$(\alpha h\nu) = B(h\nu - E_{\rm g})^2, \qquad (1)$$

where $h\nu$ is the photon energy and α is the absorption coefficient, which is obtained from optical absorption spectroscopy. The typical curve is given in Fig.1, in which the linear relationship between $(\alpha h\nu)^{1/2}$ and $h\nu$ can be identified and the Tauc gap can be estimated for the present a-SiC:H films. The results are listed in Table 1 and the carbon content x of a-$Si_{1-x}C_x$:H samples prepared was determined by X-ray photoelectron spectroscopy (XPS) by integrating the Si2p and C1s XPS signals. As expected, the carbon content x is gradually increased to about 99% with increasing gas fraction of xylene. Correspondingly, the Tauc gap is increased monotonously as the increase of the carbon content in a-SiC:H alloys and the maximum value can be as high as 3.5 eV, which is much larger than that of a-SiC:H alloys prepared by using methane and silane. The film with wide optical band gap suggests that the formation of graphite-like clusters is effectively reduced.

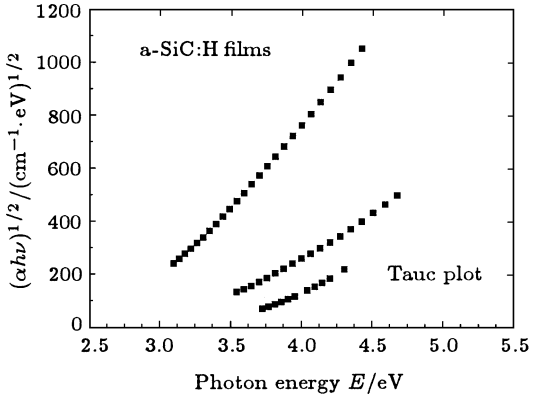


Fig.1. Tauc plots for a-SiC:H films produced by using xylene as carbon source.

It was also found that the slope of Tauc plot, $(B)^{1/2}$, is decreased with increasing optical band gap. For example, the $(B)^{1/2}$ value for film with the Tauc gap of 2.8 eV is 663.8 $\mathrm{cm}^{-1/2}\cdot\mathrm{eV}^{-1/2}$, and for film with the Tauc gap of 3.2 eV, this value decreases to 328 $\mathrm{cm}^{-1/2}\cdot\mathrm{eV}^{-1/2}$. It is considered that the $(B)^{1/2}$ value is related to the randomness of the amorphous semiconductors.[9] The low value indicates that the a-SiC:H film with high optical band gap contains a soft structure and is polymer-like.

Table 1. Tauc gap and PL peak position as a function of carbon content in a-SiC:H films.

Carbon content/%	Tauc gap Δ/eV	PL peak energy E/eV
68	2.3	1.96
78	2.8	2.06
88	3.3	2.53
95	3.5	2.76

An intense visible light emission has been observed from the present a-SiC:H alloys produced by using xylene as carbon source. Figure 2 shows the room temperature PL spectrum for the film with a Tauc gap of 3.2 eV, which was measured by using Xe lamp with wavelength of 300 nm as the excitation light. The PL peak is centered at 510 nm (2.43 eV). It was found that the PL peak energy is blue-shifted from 1.96 to 2.75 eV as the optical band gap increases from 2.3 to 3.5 eV as summarized in Table 1.

The PL shift with the optical band gap in our case can be attributed to the higher rf power density (∼200 $\mathrm{mW/cm}^2$) and the lower reaction pressure we used, compared to the previous report.[6] Under these conditions, xylene molecules are relatively fully decomposed. This results in the reduction of the aromatic structures as indicated in FT-IR spectra.[6,7] Thus, the electric and optical properties of the films are mainly determined by the σ-bonded inorganic lattice. It is unlikely that the PL is originated from the recombination through the aromatic rings molecular orbitals, as suggested by Nevin *et al.* The PL in our case is related to the transition between tail-to-tail states as in the case of a-Si:H. This results in the PL peak shifted with the Tauc gap of the films.

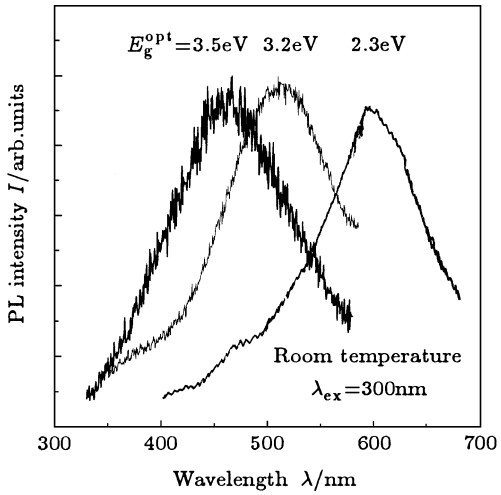


Fig.2. PL spectra for a-SiC:H films deposited by using xylene source with different optical band gap.

In order to achieve the electroluminescnce from the present a-SiC:H films, the simple structure shown in Fig.3 was fabricated. The a-Si:H and a-SiC:H films were successively deposited on the ITO coated glass

substrate in the same deposition chamber. Al electrodes with size of 1 mm in diameter were evaporated finally.

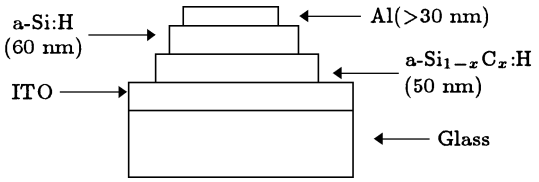


Fig.3. Sketch diagram of EL cell.

The current-voltage (I-V) characteristics curve exhibits the rectifying junction behavior and the electroluminescence could be observed both under forward and reversed bias. The EL given in this paper was measured under reversed bias and the threshold voltage is about 20 V. Figure 4 is the EL spectra for an a-SiC:H sample with optical band gap of 3.2 eV measured at room temperature under different current densities. It was found that the EL peak is located at around 600 nm (2.06 eV) and the yellowish EL light can be clearly seen by naked eyes in the dark room. The luminescence intensity is almost linearly increased with increasing applied current density due to the more electron-hole can be radiatively recombined in the film. The origin of EL needs further investigation, we tentatively attribute it to the recombination of electrons and holes, which tunnel from ITO electrode and Al electrode to the deep states of a-SiC:H films, respectively.

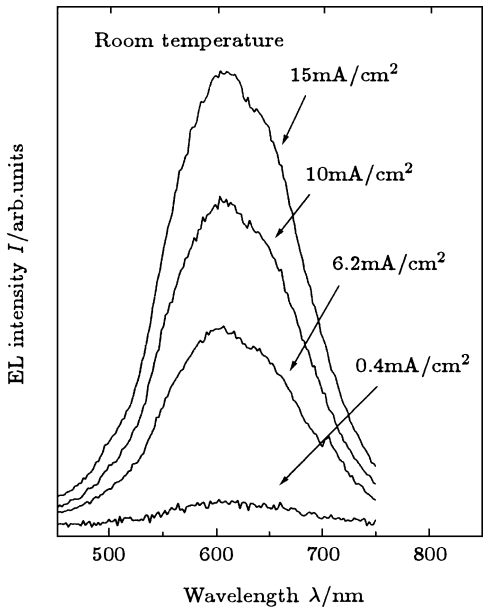


Fig.4. Room temperature EL spectra for the sample with the Tauc gap of 3.2 eV measured at different current densities.

IV. CONCLUSIONS

In summary, a series of a-SiC:H films have been fabricated by using xylene as a carbon source. It was found that the optical band gap is monotonously increased with increasing ratio of xylene to silane, and the optical band gap can be as high as 3.5 eV. For sample with high optical band gap, the structure is soft and polymer-like. Room temperature photo- luminescence was observed and electro-luminescence has been successfully obtained.

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