



# Light emission from a-Si:C:O:H films fabricated by C<sub>2</sub>F<sub>6</sub> and O<sub>2</sub>/C<sub>2</sub>F<sub>6</sub> plasma treating silicone oil liquid

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## ABSTRACT

Amorphous Si:C:O:H films were fabricated at low temperature by C<sub>2</sub>F<sub>6</sub> and O<sub>2</sub>/C<sub>2</sub>F<sub>6</sub> plasma treating silicone oil liquid. The a-Si:C:O:H films fabricated by C<sub>2</sub>F<sub>6</sub> plasma treatment exhibited white photoluminescence at room temperature, while that by O<sub>2</sub>/C<sub>2</sub>F<sub>6</sub> plasma treatment exhibited blue photoluminescence. Fourier transformed infrared spectroscopy and Raman spectroscopy studies showed that the sp<sup>3</sup> and sp<sup>2</sup> hybridized carbons, Si–C bond, Si–O bond and carbon-related defects in a-Si:C:O:H films correlated with photoluminescence. It is suggested that the blue emission at 469 nm was related to the sp<sup>3</sup> and sp<sup>2</sup> hybridized carbons, Si–C bond, carbon dangling bonds as well as Si–O short chains and small clusters, while the light emitting at 554 nm was related to the carbon-related defects.

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## 1. Introduction

Development of a light source for applications in display and lighting technologies makes the white light emission materials to be of interest. The carbon-incorporated silicon oxide is one of the promising candidates because its photoluminescence (PL) covers all visible spectral range of 350–800 nm [1]. Recently, various carbon-incorporated silicon oxide light emitting materials have been developed, including amorphous Si:C:O:H films [2], amorphous silicon oxycarbide (a-SiC<sub>x</sub>O<sub>y</sub>) films [3,4], hydrogenated amorphous silicon carbide (a-SiC:H) films [5,6], and silicon-incorporated diamond-like carbon films [7]. These films were fabricated via thermal vapor deposition [3], reactive dc-magnetron sputtering [2,6], melt-spinning [4], and plasma enhanced chemical vapor deposition (PECVD) [5,7]. During fabricating these films, a very high deposition or post-treatment temperature is needed [1–4]. For example, the deposition temperature for a-SiC<sub>x</sub>O<sub>y</sub> films grown via thermal vapor deposition was 800 °C [3], while the sintered temperature for a-SiC<sub>x</sub>O<sub>y</sub> films by melt-spinning was as high as 1300 °C [4]. Unfortunately, the high temperature steps are not desirable in some applications. Therefore, the growth of a-Si:C:O:H films at low temperature was developed, however, the oxidation treatment of a-Si:C:O:H films was still carried out at 450 °C [2]. So, developing a method to

fabricate a-Si:C:O:H light emitting materials at low temperature is very important.

The development of dual-frequency capacitively coupled plasma (DF-CCP) provides a possible method to fabricate a-Si:C:O:H films at low temperature. In the DF-CCP, the high-frequency (HF) power is used to produce high density plasma, while the low-frequency (LF) power can control the ions energy to the treated materials effectively [8–10]. If we use a DF-CCP fluorocarbon plasma to treat organosilicon liquid, a-Si:C:O:H light emitting films may be obtained. However, the related work is seldom reported. Therefore, in this work, we fabricated a-Si:C:O:H films at low temperature using C<sub>2</sub>F<sub>6</sub> and O<sub>2</sub>/C<sub>2</sub>F<sub>6</sub> dual-frequency capacitively coupled plasma treating silicone oil liquid, and investigated the photoluminescence and structural characteristics of a-Si:C:O:H films.

## 2. Experimental details

In the experiment, the silicone oil with viscous coefficient of 500 mm<sup>2</sup> s<sup>−1</sup> was used as original material. A thin layer of silicone oil liquid at quartz wafer was prepared by a spin-on method at 4500 rpm. Its thickness was about 0.5 mm. Then the samples were treated by 60 MHz (HF)/2 MHz (LF) dual-frequency capacitively coupled plasma (DF-CCP) [11]. The HF power of 165 W (Comdel, CV500) was applied to the top electrode to produce plasma. The thin layers of silicone oil liquid were placed at the bottom electrode applied the LF power of 100 W (Comdel, CX600), which controlled the ions energy to the samples. The base

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pressure and the work pressure were  $5 \times 10^{-4}$  and 50 Pa, respectively. The  $C_2F_6$  (99.99%) and  $O_2$  (99.999%) with the flow rate of 30 and 3 sccm respectively were used as discharge gases. The plasma treatment was made for 30 min. After plasma treatment, the temperature increase of quartz wafer below  $100^\circ\text{C}$  was measured with a thermometer.

The photoluminescence measurements were carried out with a JASCO FP-6500 spectrofluorometer in the wavelength range of 250–600 nm with the resolution of 1 nm. The wavelength of excited light is 220 nm. The bonding configurations of original and post-treatment silicone oils were measured by a Nicolet MagNa-IR550 Fourier transform infrared (FTIR) spectroscopy in the wavenumber range of  $400\text{--}4000\text{ cm}^{-1}$  with the resolution of  $3\text{ cm}^{-1}$ . The microstructure of silicone oil surface was observed using optical microscope attached a CCD camera. X-ray diffraction (XRD) was performed on a standard power diffractometer (Rikagu D/MAX-2000PC) using  $\text{Cu K}\alpha$  ( $\lambda=0.154051\text{ nm}$ ) radiation. The Raman spectra of the samples were recorded with JY HR800 Raman spectrometer in the Raman shift range of  $100\text{--}4000\text{ cm}^{-1}$  with the resolution of  $1\text{ cm}^{-1}$ . The laser excitation line was 514 nm.

### 3. Results and discussion

#### 3.1. Microstructure

Fig. 1(a) shows the microstructure of the original silicone oil. It can be seen that its surface is very smooth. After plasma treatment, a yellow thin solid film was obtained. By XRD analysis, the Gaussian bands between  $2\theta=7.6^\circ$  and  $16.3^\circ$  mean that the thin solid films are an amorphous structure, as shown in Fig. 2. Fig. 1(b) shows the surface microstructure of thin solid film fabricated by  $C_2F_6$  plasma treatment. A reptation condensed structure is observed [12]. This structure can be seen in some polymer dynamics, such as the boundary dynamics on the viscosity of polymer melts [13], and the molecular assembly [14]. In our work, the formation of reptation condensed structure may be related to the round movement of silicone oil liquid from bottom to top due to the ions bombardment on silicone oil liquid [15].

#### 3.2. Fourier infrared spectroscopic studies

Fig. 3(a) shows the FTIR spectrum of original silicone oil. The peaks at  $1021.86\text{ cm}^{-1}$  and  $1092.15\text{ cm}^{-1}$  with same intensity are assigned to the Si–O–Si stretching vibration. Because the silicone oil is a molecule with long Si–O chain, the Si–O absorption splits into these two peaks [16]. The peaks at  $799.13$  and  $863.86\text{ cm}^{-1}$  are assigned to the Si–C in  $\text{SiMe}_2$  and  $\text{SiMe}_3$ , the peak at  $1261.28\text{ cm}^{-1}$  is assigned to the Si–CH<sub>3</sub>, the peaks at  $2904.84$  and  $2962.56\text{ cm}^{-1}$  are assigned to the  $sp^3\text{ C-H}_2$  and  $sp^3\text{ C-H}_3$ , the peaks at  $668.98\text{ cm}^{-1}$ ,  $684.99\text{ cm}^{-1}$  and  $700.99\text{ cm}^{-1}$  are assigned to the Si–C–H, C–H and Si–O, and the peaks at  $1412.32$ ,  $1447.12$  and  $1591.19\text{ cm}^{-1}$  are assigned to the Si–CH=CH<sub>2</sub> and  $sp^2\text{ C=C}$ . The Si–O bond is the main chain of silicone oil molecule, the Si–CH<sub>3</sub>, Si–C and Si–C–H are the side chains, and the C–H<sub>3</sub>, C–H<sub>2</sub> and C–H are the terminal groups. The Si–CH=CH<sub>2</sub> and C=C may be from the impurity in silicone oil liquid.

Fig. 3(b) shows the FTIR spectrum of silicone oil treated by  $C_2F_6$  plasma. The C–H ( $684.99\text{ cm}^{-1}$ ) disappears. The red-shift of Si–O–Si vibration from  $1021.86$  to  $1013.50\text{ cm}^{-1}$  takes place and its intensity decreases. Additional, the Si–C–H ( $668.98\text{ cm}^{-1}$ ), Si–C in  $\text{SiMe}_2$  ( $799.13\text{ cm}^{-1}$ ), Si–O–Si ( $1092.15\text{ cm}^{-1}$ ), Si–CH<sub>3</sub> ( $1261.28\text{ cm}^{-1}$ ), and  $sp^3\text{ C-H}_3$  ( $2962.56\text{ cm}^{-1}$ ) all decrease.

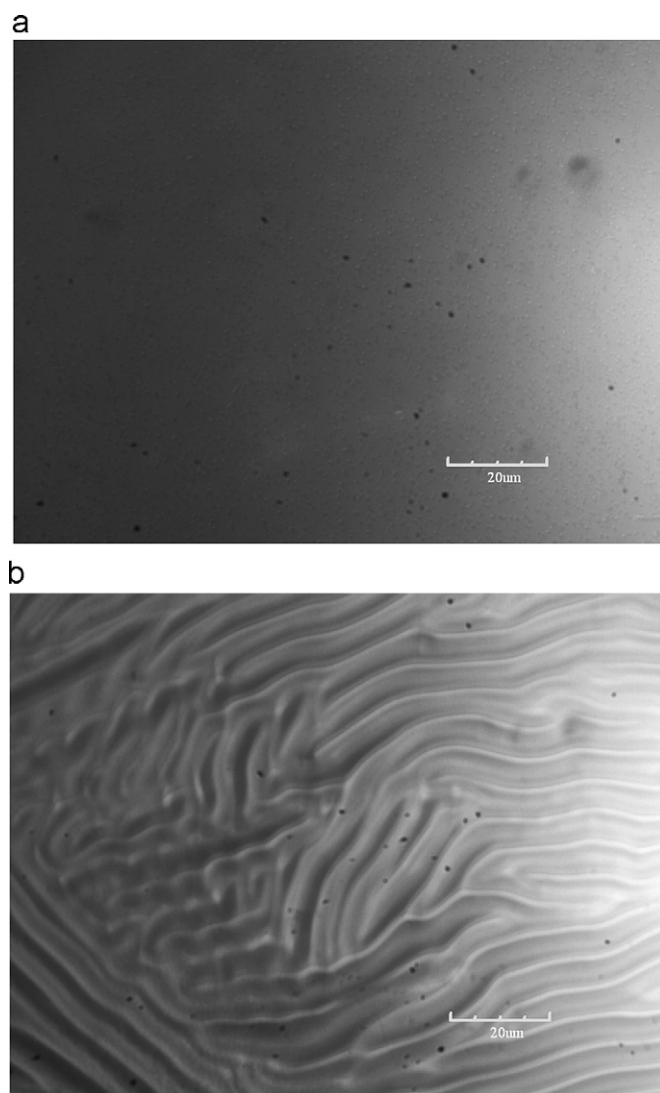


Fig. 1. Microstructure of (a) original silicone oil and (b) the silicone oil treated by  $C_2F_6$  plasma.

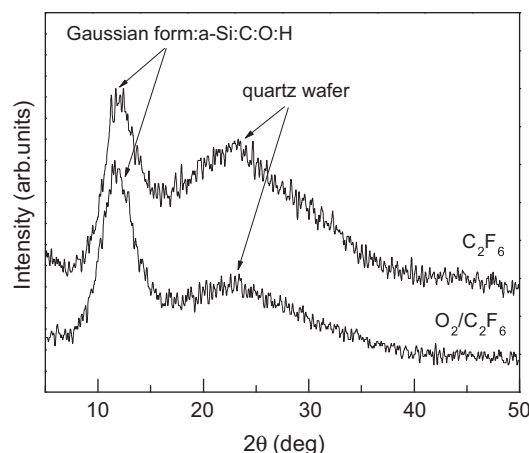


Fig. 2. XRD spectra of the silicone oils treated by  $C_2F_6$  and  $O_2/C_2F_6$  plasmas.

No  $\text{CF}_x$ -related bonding configuration is seen. The intensity decrease of Si–O peaks means the broken of long Si–O chain and the formation of short Si–O chains in the film [16], while the intensity decrease of  $sp^3\text{ C-H}_2$  and  $sp^3\text{ C-H}_3$  peaks means the

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