Precipitation of Nanocrystallites in Amorphous Silicon Carbide Films by Low Temperature Catalytic CVD

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Amorphous silicon carbide (a-Si:C) films are of great interest for applications to optoelectronic devices and photovoltaics. The band gap of this material can be tailored in a wide range by changing the mixing ratio of the source gases during the film deposition process. Nanocrystalline particles embedded in the amorphous matrix of the film can modify the band structure, leading to unique optical characteristics. Deposition of these films on plastic substrates can also open up new possibilities of manifesting various wearable devices.

In this study, a-Si:C films were prepared at a substrate temperature of 200°C by catalytic chemical vapor deposition (Cat-CVD) technique. A source gas mixture of silane (SiH_4), methane (CH_4) and hydrogen (H_2) were introduced to the reactor through an array of tungsten filament heated to 1750°C. Self-heating of the substrate by radiative heat from the filament was suppressed by using a cooling system underneath the substrate holder. Flow rates of SiH_4 and H_2 were fixed at 2 sccm and 10 sccm, respectively, and that of CH_4 was employed as the main process variable.

As shown in Fig. 1, transmission spectra of a-Si:C films exhibited variation with CH_4 flow rates. The shoulder appearing at the wavelengths of 450-500 nm corresponded to the band-to-band transition of a-Si:C. The secondary absorption was observed in the range of 640-680 nm and its position shifted toward the low wavelength region as the CH_4 flow rate increases. This range of wavelength corresponded to absorption in nanocrystalline silicon. Fourier transformation infrared absorption (FTIR) analyses showed a high intensity of Si-C bonding, indicating that the matrix consisted of a-Si:C. As the CH_4 flow rate in the source gas increased, Si-CH_3 and Si-H_2 bonding concentrations increased. These bonds are mainly formed on the surface of second phase inclusions. High resolution transmission electron microscopy revealed a columnar growth of an interfacial layer as thick as 40 nm. Selective area diffraction pattern indicated that this layer is amorphous. Energy dispersive X-ray scan (EDS) showed that silicon counts dropped considerably in this region. This result indicated that adsorption of carbon on the growing surface was preferred in this early stage, and the high concentration of Si-CH_3 bonds found in the FTIR spectra could be attributed to the columnar structure of the interfacial layer. On top of this interfacial layer was found a homogeneous a-Si:C layer. The EDS showed that this layer was rich in silicon. Diffraction spots were found in this layer, suggesting the existence of silicon nanocrystallites of a few nanometers in diameter which might be responsible for the optical absorption in the range of 640-680 nm.

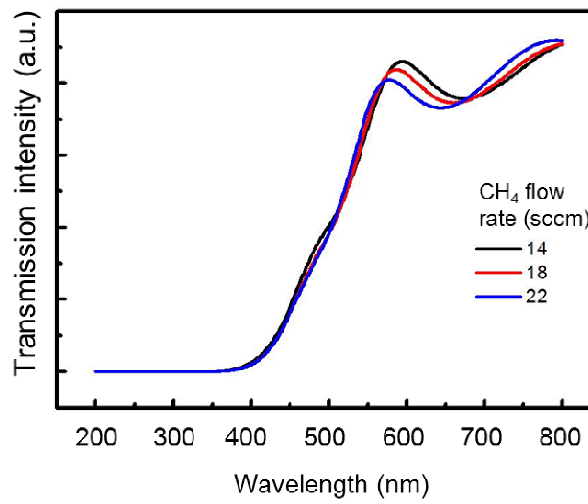

Fig 1 Change in transmission spectra of low temperature a-Si:C films prepared with different CH_4 flow rate in the source gas supply.