

Optical Characteristic of Al Doped ZnO Film Deposited by Radio Frequency Magnetron Sputtering on SiOC

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Abstract—The aluminum doped zinc oxide films were deposited on SiOC/p-Si wafer by an RF magnetron sputtering system, varying the deposition parameters such as radio frequency power and argon flow rate. The optical properties of AZO thin films on SiOC/Si wafer were studied by PL (photoluminescence) and the roughness to assess the correlation between the uniformity of PL intensity and opto-electronic characteristics. To research the quality of AZO film on SiOC/Si wafer, AZO films were prepared on SiOC/Si films at different Ar gas flow rates. The emission peaks covering wide visible range was observed by increasing RF power and Ar gas flow rate owing to the quantum energies based on the diversity of the defects. AZO/SiOC/Si films showed the weak blue emission owing to the free exciton recombination at the interfacial between SiOC and AZO films, and strong green and red emissions due to the defective structure. The green emission is associated with oxygen vacancies which are electron donors. Transmittance was increased at AZO grown on SiOC film. The PL spectra of AZO/SiOC/Si film prepared by using sputtering method covered the entire visible spectrum, resulting in white light emission, which is useful as the white light sources such as LED or solar-cell. AZO as transparent conductive oxide improved on amorphous based SiOC film.

Index Terms—AZO, TCO, SiOC, PL, oxide semiconductor.

I. INTRODUCTION

For devices applications, the growth of high quality ZnO with a smooth surface morphology and a high crystalline ordering, as well as high emission efficiency, is essential to assure a high performance and long lifetime devices. ZnO has been studied as a semi conducting material and has attracted attention due to improvements in deposition techniques that have made it possible to make high-quality ZnO thin films.¹⁻⁴ ZnO is a II-VI compound semiconductor with a wide band gap of 3.37 eV. If the ZnO semiconductor characteristics can be utilized, high-power electronic devices, such as select-transistors in individual pixels of an active-matrix liquid-crystal display, might be possible [1-3]. Furthermore, ZnO is one of a few oxides that can be grown as a crystalline material at relatively low deposition temperatures. The sputter deposition of ZnO film should be performed at low temperature because of the problem of the deformation due to the degradation of the film properties. Various techniques have been used in the deposition of TCO film such as ITO (Tin doped indium oxide), AZO (Al doped Zinc oxide), and

IGZO (Indium gallium Zinc oxide), beings the most commonly used radio frequently magnetron sputter, that is consider to be one of the most favorable deposition due to its high reproducibility, low temperature process and smooth surface with good uniformity. Al doped ZnO (AZO) films have attractive much attention because of their comparative high optical transmittance and low electrical resistivity with respect to other TCOs widely used such as Zinc oxide (ZnO) and Tin-doped Indium Oxide (ITO) films[4-7]. In order to achieve the enhancement of white light emission and high transparent conductivity, ZnO has been widely investigated on various substrates or porous templates. The crystal quality of ZnO thin film on Si depends on interface properties between ZnO and Si, which induces the large lattice mismatch between Si and ZnO and the formation of amorphous SiO_x layers. And the grain boundaries observed in polycrystals reduce emission efficiency because they are scattering source for photons and also may be a path for leakage current. SiOC film is promising ILD materials as low-k materials, which replaces the silicon dioxide thin film ($k \approx 4.0$) as an insulator for silicon based devices. Especially, low polarization SiOC thin films (LP-SiOC) by the chemical vapor deposition (CVD) was reported that have high degree of amorphous structure and the low dielectric constant of $k \approx 2.0$ due to the decreasing the polarization by the ionization between opposite dipole group[8-10].

In this paper, in order to improve the uniformity and high degree crystallization, AZO thin films were grown on SiOC film by RF magnetron sputter, and the mechanical-chemical properties of the AZO thin films on SiOC film were investigated by atomic force microscopy and photo luminance.

II. EXPERIMENT DETAILS

The AZO ceramic target (ZnO: Al₂O₃, 98%:2% wt) were supplied by the sputtering RF power of 200W. The flow rate of the argon (99.9999%) was controlled by mass flow controller (MFC) from 5~30 sccm for 15 min. The substrate for AZO thin film was n-type Si wafers controlled by RF magnetron sputter at a pressure of 0.01 Torr in argon atmosphere. The target to substrate distance was kept at 100 mm and the base pressure was 4.5×10^{-5} Pa and the working pressure of the chamber with argon gas was $1.2 \sim 1.4 \times 10^{-3}$ Torr. The substrate temperature was kept room temperature.

SiOC films were formed by inductively coupled plasma chemical-vapor deposition (ICP-CVD) using the gas flow rates of 5sccm:5sccm with oxygen and dimethyl dimethoxy silane (DMDMOS) carried by Ar gas. Before leading the substrate in CVD chamber, they were ultrasonically cleaned by using acetone, hydrofluoric acid solution and deionized water, and then dried by blowing nitrogen over them. The DMDMOS was vaporized and carried by argon gas at 35 °C with a thermostatic bubbler. The base pressure of the mixture was kept at 3 Torr and the rf power was 800 W in each experiment. In order to achieve the growth of AZO thin film depending on substrates of n-Si and SiOC/n-Si, AZO films on n-Si and SiOC/n-Si were deposited with the same RF parameter, gas flow rate and on a substrate temperature at room temperature. The roughness and images were observed by the atomic force microscopy (AFM, S.I.S., GmbH at Cheongju University), PL spectra were obtained by the UV-visible spectrometer with xenon lamp as a light source at High Technology Components & Materials Research Center, Busan, South Korea.

III. RESULT AND DISCUSSION

In order to optimize the sputtering conditions of the AZO films deposited, it has to be taken into account that the high energy particles reaching the surface of the substrate grown on the substrate. The PL spectra of Si wafer and AZO/Si film was analyzed in Fig. 1.

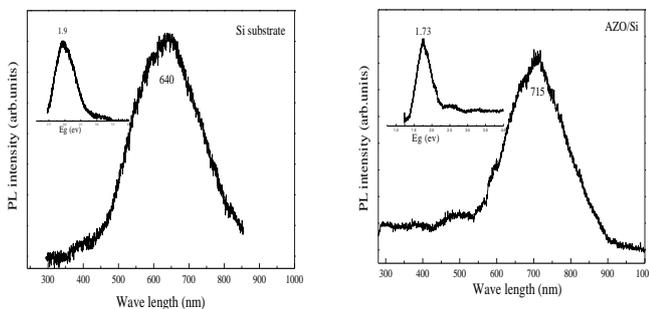


Fig. 1. PL spectra and energy gap, (a) Si wafer, (b) AZO/Si thin film.

The energy gap of Si wafer in Fig. 1(a) was 1.9 eV and PL signal peak centered at 640 nm was broad. After AZO deposition by using the RF magnetron sputtering system, the central peak of PL signal moved to higher wavelength, 715 nm, and the energy gap was 1.73 eV, respectively. The strong red emission was due to a radiative recombination of excitons in the surface region in Si. The weak green and orange-red emissions in the visible range can be attributed to native defects in AZO and to an oxygen bond in Si wafer. In view of

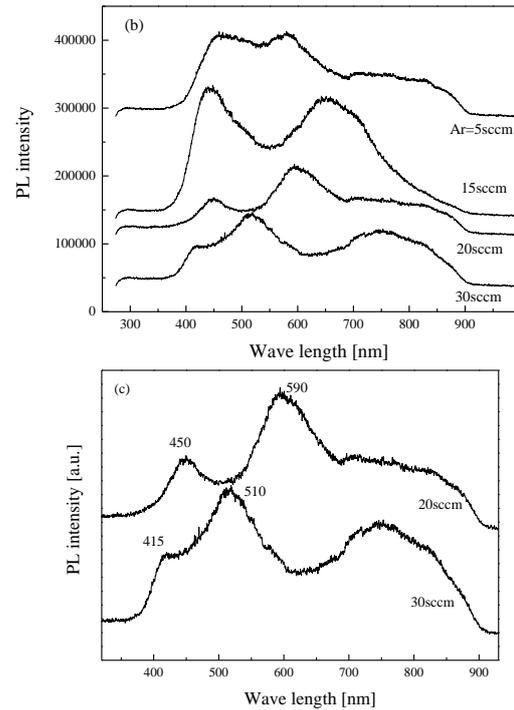
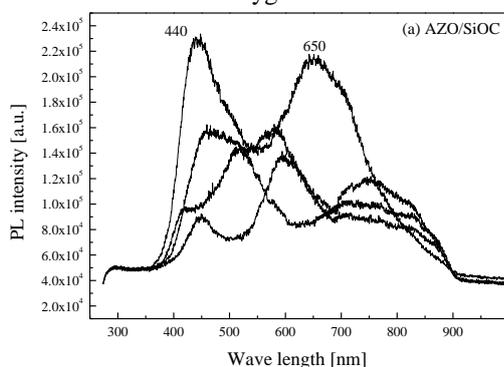


Fig. 2. PL spectra of the AZO/SiOC films grown on p type Si wafer depending on Ar atmosphere of various gas flow rates, (a) comparison of the intensities, (b) comparison of the spectra patterns, (c) blue shift in the first peaks near 450 nm shown in samples with argon gas of 20 and 30 sccm.

Fig. 2(a), the intensity of PL spectra of AZO/SiOC film deposited at 15 sccm is very strong at 440 and 650 nm, especially. As the flow rate of argon gas increases, the PL spectrum shows the strong red emission, and the intensity near 750~800 nm of AZO/SiOC film deposited at 30 sccm is higher than that of the others. Increasing the relative intensities of the yellow and green-red emissions at flow rate 20 and 30 sccm, is due to the decrease of the quantum energy. The film at 15 sccm induces two signals with strong intensity in PL spectra.

Figure 2(c) signifies the blue shift of the first peaks near 450 nm in the samples of argon flow rate 20 and 30 sccm, and the PL intensity in blue region of the sample at 30 sccm increased compared with that of the sample at 20 sccm. This result indicates the increase of the quantum energy due to the blue chemical shift under a condition of risings the gas flow rate from 20 to 30 sccm. The sample at 20 sccm involves the emission peak at 590 nm, which means that there are electron acceptor defects in AZO thin film. But the one at 30 sccm has the emission peak at 510 nm associated with the electron donor defects such as oxygen vacancies and Zn ions which contribute to high conductivity. Especially, AZO/SiOC /Si thin film prepared at 30 sccm is useful in the field of Display or solar cells with high efficiency as transparent conductive oxide because the film shows the pattern of wide band emissions in visible range involving the blue in a near band edge, green at 510 nm, and the red at 700~900 nm, by emitting the white light luminescence with high quality.

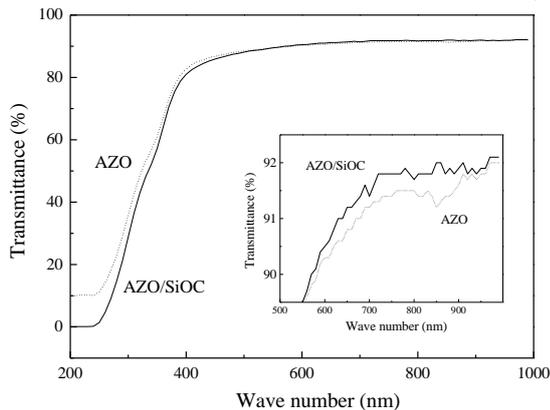


Fig. 3. Transmittance of AZO/SiOC and AZO thin films grown on Si substrate.

Figure 3 was the transmittance of AZO/SiOC and AZO on bare Si wafer. The transmittance of AZO/SiOC increased in the visible region in compared with that of AZO on Si wafer. These spectra are referred to air, and the absorption of light in the glass substrates was not subtracted from the transmittance spectra. All the samples were found to be higher transparent with an average transmittance over 90% in the visible range. The transmittance of AZO/SiOC thin film on Si wafer increased to 92% at 700 nm wavelength because of increasing the electron donor defects such as oxygen vacancies and Zn ions at the interface between AZO and SiOC film. It might to be attributed to the deep level emission by the defects and the decrease of crystallinity by Al dopants in the ZnO lattice. However these effects improved the transmittance in the visible region. Consequently, it was obtained that Al doped ZnO is suitable for the transparent conductive oxide thin film in semiconductor devices such as display, solar cell or LED.

IV. CONCLUSION

The optical properties of AZO thin film prepared on SiOC film by RF magnetron sputtering were studied, and SiOC film was prepared by the chemical vapor deposition. The optical properties of AZO films were considerably improved by optimizing the process conditions. To show optical properties as the candidate of white light luminance, broad pattern of PL spectrum should be resulted with high transmittance in the visible region. A white light luminescence resulted by blue, green and red emission peaks was observed from the AZO/SiOC/p-Si thin film. White light emission across the visible light region is attributed to the diversity of the defects, and to the transmittance and thickness of the film when AZO film is deposited on the surface of Si based substrate through the siloxene structure. The result that the AZO/SiOC thin film emits the white light luminescence indicates that the AZO thin film deposited by sputtering at room temperature can be used in various types of potential applications such as solid white light source, solar cell, and LED device with high efficiency, etc. as a transparent conductive oxide thin film.

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