

Low temperature growth of ZnO thin film on Si(100) substrates by metal organic chemical vapor deposition

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In recent years, wide band-gap semiconductor materials have attracted a great deal of attention for use in blue light-emitting and short-wavelength diodes. Additionally, due to its high conductance, chemical and thermal stability, and high piezoelectric coupling coefficient, ZnO is also used for piezoelectric devices, such as surface acoustic wave (SAW) devices [1] and bulk acoustic devices [2]. Various deposition techniques, including sputtering [3, 4], pulsed laser deposition (PLD) [5, 6], ion beam deposition [7], chemical vapor deposition (CVD) [8, 9], atomic layer deposition (ALD) [10], metal-organic chemical vapor deposition (MOCVD) [11, 12], and molecular beam epitaxy (MBE) [13] have been employed for the growth of ZnO films. However, MOCVD has an advantage in achieving devices in commercial level since high deposition rate and high-quality film is attainable, especially at low pressure. High quality ZnO films grown on Si substrate pave the way from integration of devices with Si IC technology. Also, amorphous substrates such as SiO₂ and glass substrate have obvious technological advantages and potential applications [14]. Although most researchers have grown ZnO films on sapphire substrates, there are not many reports on growing ZnO thin films on Si or Si-based materials by MOCVD technique. Furthermore, there are rare reports on the MOCVD growth of ZnO at low temperatures below 300 °C.

In this work, we investigate the growth of ZnO thin films on silicon substrate at the temperature range of 100–200 °C by employing the MOCVD technique. Since the material properties strongly affect the performance of a device, the influence of substrate temperature on the structural properties of the samples are presented.

The ZnO films were deposited on p-type Si(100) substrates by MOCVD system using Zn(C₂H₅)₂ (99.9999% purity DEZn (diethylzinc)) and O₂ (99.999% purity). Fig. 1 shows a schematic diagram of the MOCVD reactor used in our experiments. Before loading into the reactor, the substrate was cleaned in acetone for 10 min, HF (20:1) for 1 min and then rinsed by deionized water for 1 min. High-purity Ar was passed through the DEZn bubbler and saturated with DEZn vapor to the reactor. For MOCVD growth of ZnO films, the gas phase reaction will result in particle formation, which will degrade the ZnO film quality. In order to minimize the gas phase reaction, Zn(C₂H₅)₂

and O₂ are introduced into the reactor separately and mixed just before the inside of chamber. Zn(C₂H₅)₂ bubbler was maintained at the temperature of –2 °C. Here, the ratio of Ar to O₂ gas flow rates was set to 2. The growth temperature ranged from 100 to 200 °C at a pressure of 5.0×10^{-1} Torr and deposition was carried out for 30 min. The structural characteristics of the films were analyzed by X-ray diffraction (XRD) using CuKα1 radiation ($\lambda = 0.15405$ nm) and by scanning electron microscopy (SEM: Hitachi S-4200).

Fig. 2 shows XRD patterns of ZnO thin films on Si(100) substrates in the temperature range of 100–200 °C. The θ - 2θ scan data of ZnO films exhibits a strong 2θ peaks at 34.53° in the sample grown at 200 °C, corresponding to the (002) peaks of ZnO. The observation of the strong (002) peak indicates that the film is grown with a c-axis orientation. The film has other peaks such as (101), (100), and (102), etc., corresponding to granular structure of the film. Furthermore, investigation of the XRD patterns of the ZnO film grown on Si(100) substrate indicates that full-width at half-maximum (FWHM) of the (002) diffraction peak is about 0.44° at a growth temperature of 200 °C. On the other hand, at lower temperatures in the range of 100–150 °C, the relative intensity of the (002)

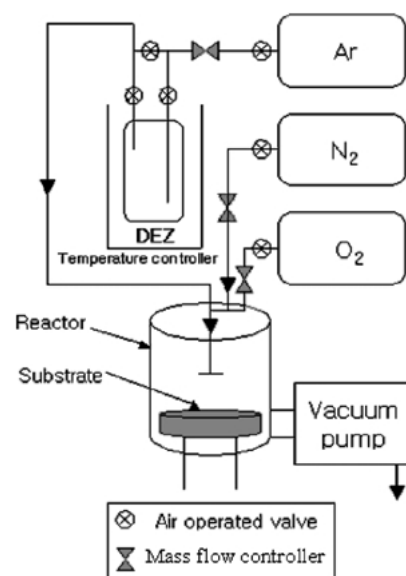


Figure 1 Schematic diagram of MOCVD reactor.

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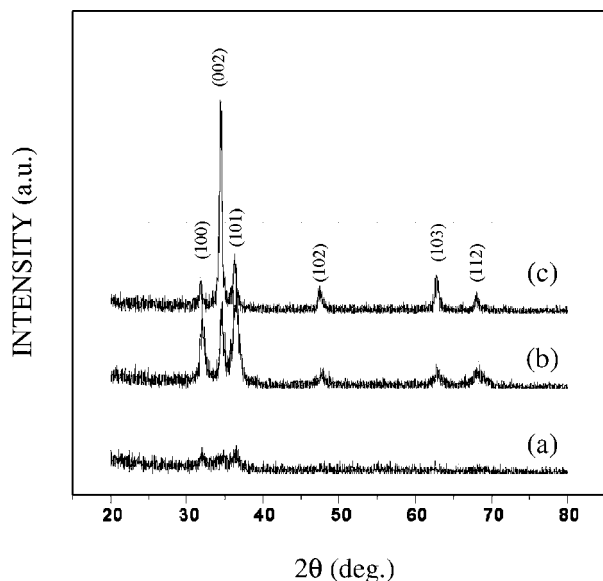


Figure 2 XRD patterns of ZnO thin films on Si(100) substrate with growth temperature of (a) 100 °C, (b) 150 °C, and (c) 200 °C.

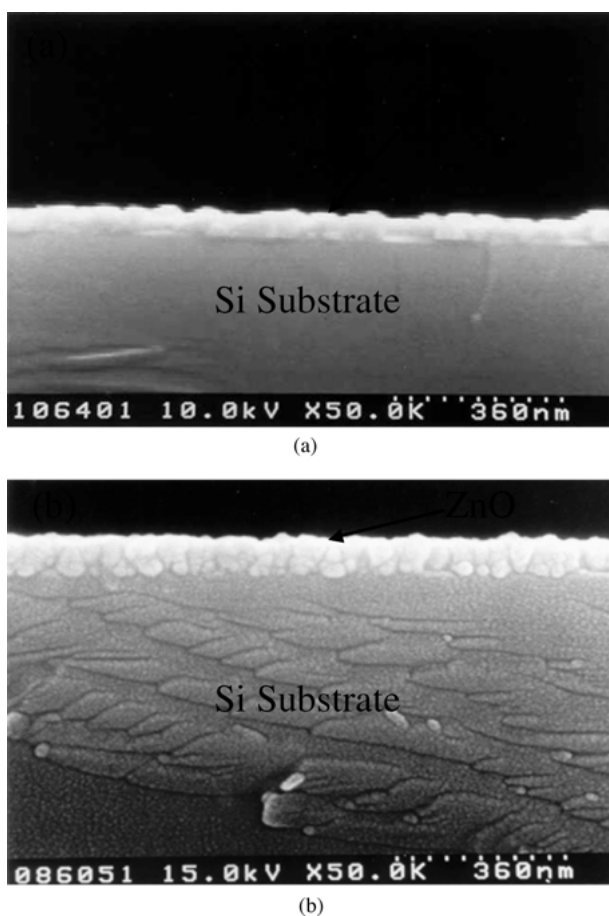


Figure 3 Cross-sectional view of ZnO film grown on Si(100) substrate at the growth temperature of (a) 100 °C and (b) 200 °C.

diffraction peak compared to the neighboring (100) and (101) peaks is lower than the relative intensity at 200 °C. We surmise that the degree of c-axis orientation in ZnO film decreases by decreasing the growth temperature.

Fig. 3a and b show the SEM images of ZnO thin films grown on Si(100) substrate at 100 °C and 200 °C,

respectively. The thickness of ZnO layers are measured to be 60 nm and 84 nm, respectively. They show that the ZnO film structure grown at 200 °C consists of some columnar-structured grains, representing c-axis oriented grains. However, the ZnO film structure grown at 100 °C does not clearly show the columnar structured grains. They agree with XRD data. Although we cannot explain the growth mechanism at this moment, further study is underway to understand this observation.

In conclusion, we demonstrate the growth of ZnO thin films on Si(100) substrate at low temperature of 100–200 °C, using the MOCVD reactor. The c-axis orientation based on XRD analysis improves with increasing substrate temperature. X-ray diffraction reveals that the ZnO films are highly c-axis-oriented and the line width of ZnO(002) peak is significantly small and the full width at half maximum (FWHM) of 0.44° was achieved at 200 °C. We also reveal that the growth of ZnO thin film on silicon substrate is achievable even at a low temperature of 100 °C. Growth temperature of high quality ZnO films is significantly lowered using MOCVD, showing light on the potential application of ZnO film on electronic and optoelectronic devices.

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