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Lower temperature growth of single phase MgZnO films in all Mg content range

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A B S T R A C T

MgZnO films are grown on (0001) sapphire substrates by pulsed laser deposition. The crystal structure and optical properties of the MgZnO films have been systematically investigated with the methods of energy dispersive spectroscopy (EDS), X-ray diffraction and spectrophotometer. The EDS results show that the MgZnO films with different Mg contents can be prepared by adjusting the Mg content in the target and substrate temperature. Single phase MgZnO films are successfully obtained in all Mg content. The structural transition from hexagonal to cubic phase has been observed at the Mg content around 0.4. Optical analysis indicates that the bandgap of the MgZnO films can be tailored by controlling the Mg content in the films.

1. Introduction

ZnO is a II–VI wide bandgap (3.37 eV) semiconductor with a large exciton binding energy of 60 meV. It is expected that alloying ZnO with MgO can tune the bandgap from 3.37 eV to 7.8 eV [1,2]. Therefore, MgZnO alloy material has received much attention due to their potential applications in the short-wavelength optoelectronic devices such as light-emitting diodes (LEDs) and laser diodes [3–6].

Since the ionic radius of Mg^{2+} (0.057 nm) is similar to that of Zn^{2+} (0.060 nm), there can be some replacement in either structure without changing the original structure when alloying [7]. However, there is large crystal structure dissimilarity between wurtzite hexagonal ZnO and rock-salt-cubic MgO, which leads to unstable phase mixing. In the phase diagram of the ZnO–MgO binary system, the thermodynamic solubility limit of MgO in ZnO is only 4 at.% and MgO allows a maximum of 56 at.% ZnO solubility at 1600 °C [8]. Vashaei et al. [9] have tried to use plasma-assisted molecular-beam epitaxy (MBE) for growing MgZnO films over a wide Mg composition range from 0 to 0.97 and have found that phase separation occurs in MgZnO films with Mg content from 0.34 to 0.65. Similar results have been reported for the MgZnO films obtained by sputtering growth technique [10,11]. Pulsed laser deposition (PLD) is an effective growth method for fabricating such metastable phase films due to the relative high kinetic energies that the ablated species have [12–14]. Using this method, Ohtomo et al. have succeeded in growing single phase wurtzite MgZnO films with Mg content up to 0.33 [15]. In this paper, we report on the successful growth of single phase MgZnO films without phase separation in all Mg content range by PLD.

2. Experiment

A series of MgZnO films were fabricated by PLD using a KrF laser source (λ = 248 nm) on (0001) sapphire substrates. MgZnO bulks with different Mg content were used as targets. Before growth, the sapphire substrates were cleaned in organic solvents ultrasonically, chemically etched in a hot H_2SO_4:H_3PO_4 (3:1) solution, then rinsed in deionized water. The pulsed laser with a frequency of 2 Hz was irradiated and the distance between targets and substrates was about 30 mm. Pure oxygen gas (99.999%) was introduced through mass flow controllers after the growth chamber was evacuated below 5×10^{-6} Pa. The oxygen pressure during the growth was maintained at 1×10^{-1} Pa while the substrate temperatures were kept at 400 °C or 500 °C. The deposition time was 40 min for all samples.

The element contents in the prepared MgZnO films were determined by energy dispersive X-ray spectroscopy (EDS). The thicknesses of the MgZnO films were measured by using a surface step profile analyzer. The crystal structures of the layers were examined by conventional θ–2θ X-ray diffraction (XRD) using Cu Kα emission line. The optical transmission spectra were measured with a spectrophotometer at room temperature.

3. Results and discussion

Fig. 1(a) and (b) show the EDS of MgZnO films grown using various Mg content in the targets (x) at substrate temperature of 400 °C and 500 °C, respectively, From the spectra, elements of O, Zn, Mg, and Al are observed. The peak related to Al is attributed to the sapphire substrate due to the thickness of the films were smaller than 600 nm. It is obvious that the intensity ratio of Mg/

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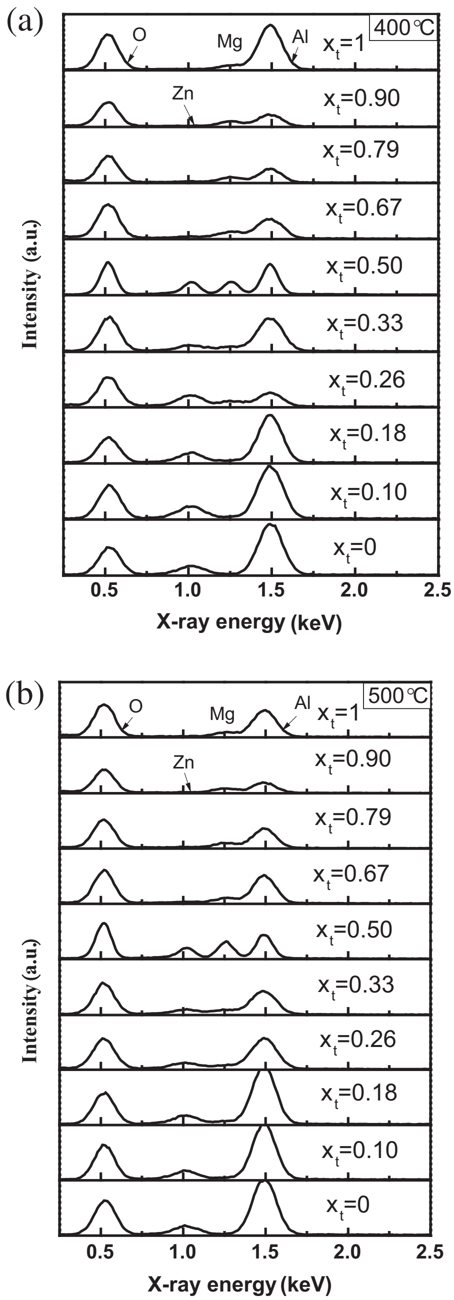

Fig. 1. EDS of MgZnO films grown at substrate temperatures of (a) 400 °C and (b) 500 °C by using MgZnO targets with various Mg content *x*.

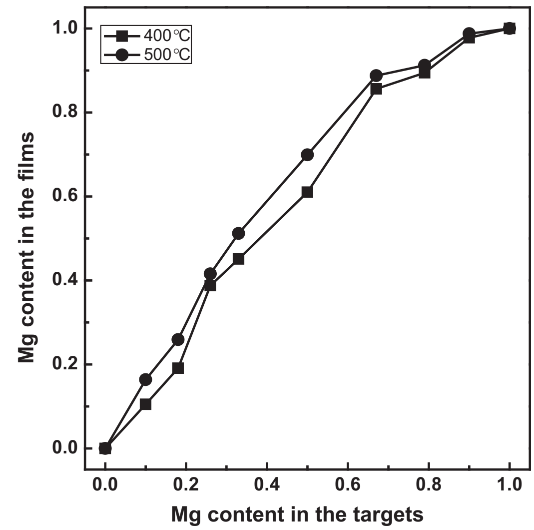

Fig. 2. Dependence of the Mg content in the obtained MgZnO films grown at the substrate temperatures of 400 °C and 500 °C on the Mg content in the MgZnO targets.

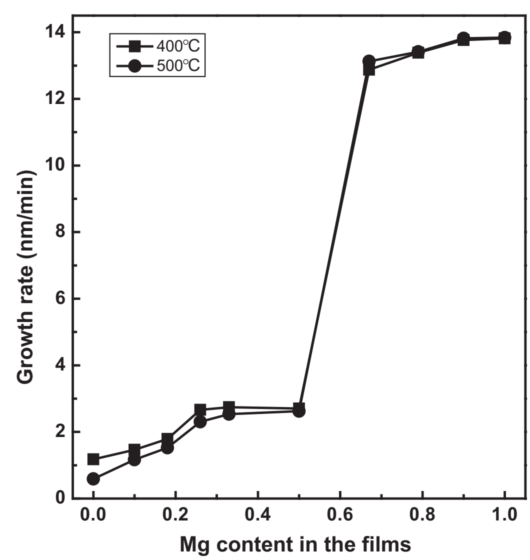

Fig. 3. Dependence of the growth rate of MgZnO films grown at the substrate temperatures of 400 °C and 500 °C on the Mg content in the MgZnO targets.

Zn in the MgZnO films increases with the increase of Mg content in the targets both for substrate temperatures of 400 and 500 °C. Fig. 2 presents the dependence of Mg content *x* in MgZnO films obtained from the EDS spectra on Mg content in the MgZnO targets. The Mg contents in the films were also confirmed by using Mg 2p and Zn 2p core level peak areas of X-ray photoelectron spectroscopy spectra after applying atomic sensitivity factors. We see that the Mg content in the films increases almost linearly with the increase of Mg content in the targets, suggesting the composition of MgZnO films can be controlled by adjusting Mg content in the targets. From Fig. 2, it is clear that the Mg content in the MgZnO films grown at substrate temperature of 400 °C is smaller than that of MgZnO films grown at substrate temperature of 500 °C for same Mg content in the MgZnO targets, indicating that substrate temperature is also an important parameter to affect the composition of MgZnO films in the PLD process.

Choopun et al. [16] have fabricated MgZnO films with Mg content ranging from 0.5 to 1 with PLD by controlling the substrate temperature from room temperature to 750 °C using one MgZnO target with Mg content of 0.5. This phenomenon can be explained by the difference of vapor pressure between Mg and Zn species. Zn species have a higher vapor pressure and can be desorbed more easily than that of Mg species at same growth temperature which results in the more Mg enriched films at high growth temperatures. In order to verify this phenomenon, we investigated the growth rate of MgZnO films at the substrate temperature 400 °C and 500 °C as shown in Fig. 3. The growth rate of the MgZnO films grown at the substrate temperature 500 °C is clearly lower than that at 400 °C for a given MgZnO target in the low Mg content region due to the Zn desorption. However, the growth rates for both of substrate temperature 400 °C and 500 °C are almost same in the high Mg content region. The results are consistent with that reported by Choopun et al. [16].