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실험계획법을 이용한 GZO 투명전극 성장의 공정 최적화

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Processing optimization of Ga-doped ZnO for transparent electrode application using DOE

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Abstract -Microstructure and electrical properties of Ga-doped ZnO (GZO) films grown on Al_2O_3 templates by Pulsed Laser Deposition (PLD) are investigated utilizing X-ray diffraction method and Hall measurement, respectively. In order to determine the optimized operating condition of the PLD, statistical design of experiment (DOE) is employed. It provides the systematic and efficient methodology for characterization and modeling of PLD processing with a relatively small number of experiments. The most optimized recipe of the process factors is obtained by response optimizer in Minitab.

1. Introduction

Transparent conducting oxide (TCO) films are currently of great commercial and scientific importance for applications in flat panel displays, electrochromic windows, electro-optical devices, gas sensors, and solar cells. To date, tin-doped indium oxide (In_2O_3/Sn, ITO) has been the most popular commercial TCO material due to its excellent characteristics such as low DC electrical resistivity ($\sim 10^{-4}\Omega cm$) and high transmittance in visible region (>80%). ITO which has been the workhorse of the TCO industry for a long time, poses a serious problem due to its high cost, limited supply, and poor chemical stability. thus, there is an urgent need to develop alternative TCO materials with similar or better properties. ZnO doped with group III elements (e.g.,Al, Ga) exhibits electrical and optical properties close to that of ITO and is an attractive candidate for TCO applications because of its superior stability in hydrogen containing atmosphere, benign nature, and relatively inexpensive supply. Then microstructural characteristics and properties of GZO films for transparent electrode deposited on Al_2O_3 by PLD. Processing variables of the $Zn_{0.95}Ga_{0.05}O$ film, which was deposited at 300~500 °C and 200~500mTorr showed predominant 0001 orientation with a metallic behavior and a resistivity of $1.82*10^{-3}\Omega cm$ at 300°C. Low resistivity of the GZO films has been explained in terms of optimal combination of carrier concentration and minimized scattering, and is correlated with the microstructure and the deposition parameters. Using DOE in this paper investigates applying to determine the effect to process optimization.

2. Experiments and DOE

There are four controllable factors in DOE. GZO thin films were deposited at different condition using PLD with an Nd:YAG laser (third-harmonic 355nm Quantel Brilliant Q-switched Nd:Yttritium Aluminum Garnet laser). The laser repetition rate was 5 Hz. The deposition conditions were configured as followed: base vacuum in the chamber was lower than 5×10^{-5}Torr, the target rotated at 5rpm to preclude pit formation and to ensure uniform ablation of the target, and deposition time was maintained for 10 minutes. The structural properties of the samples were investigated by XRD, where a Ni-filtered Cu $K\alpha$ (l= 1.54056Å) source was used, and XRD patterns in $2\theta-\omega$ scan mode were measured with an interval of 0.02°. The electrical properties of GZO films were investigated by Hall measurement system in the van der Pauw configuration in a magnetic field (B=1T) at RT.

A fractional factorial design includes selected combinations of factors and levels. It is a carefully prescribed and representative subset of a full factorial design. The advantage in this method is the number of experiment half of the full factorial design. By this method, the number of experiments can be reduce in half. The number of fractional factorial experiment is determined by

$$TC_2 = K^{n-1} \quad (1)$$

where TC_2 is the number of fractional factorial experiment, K is the number of levels, and n is the number of factors.[1] In this paper, four input factors with two levels were considered: gas pressure of oxygen, substrate temperature, target-substrate distance and energy density of laser. The ranges of each factor are sown in table 1.

Table 1. The parameters of PLD processing

Parameter	Range
gas pressure	200-500mTorr
substrate temperature	300-500°C
T-S distance	4-6cm
energy density	1.39-1.54J/cm^2

3. Results and discussions

Structural properties is studied by X-ray diffraction of GZO/Al_2O_3 confirmed epitaxial growth of the (0001) orientated. The XRD spectra of these films as shown in Fig. 1 show the dominant (002) peak, which shows that the films are highly textured along the c axis and aligned with the (006) peak of sapphire. The (002) peak positions of ZnO was observed in all samples This result shows that there is no significant change in orientation of the films according to range of factors. GZO films deposited on a sapphire utilizing pulsed laser deposition method was also analyzed by DOE. According to the results of experiments in Fig. 2(a), the effect of substrate temperature and T-S distance are more sensitive on resistivity. Comparatively in Fig 2(b), (c) the effect is more sensitive on mobility and bulk concentration. According to the results of our experiments, substrate temperature and T-S distance is a very critical factor

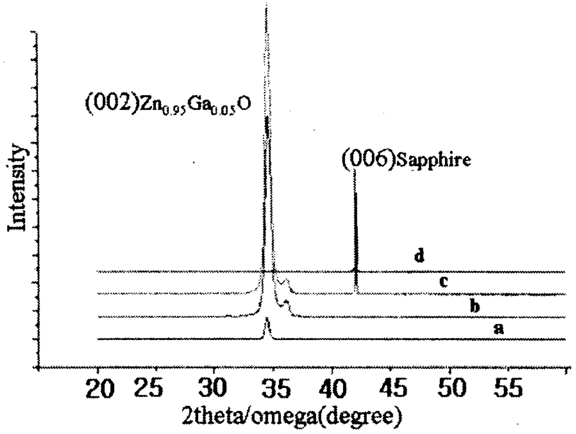

Fig 1. X-ray diffraction patterns of GZO films : (a) 500mTorr, 500℃, 1.39J/cm2, 4cm (b) 500mTorr, 500℃, 1.54J/cm2, 4cm (c) 500mTorr, 200℃, 1.54J/cm2, 6cm (d) 500mTorr, 500℃, 1.54J/cm2, 6cm

for deposition of GZO films grown by PLD. Fig 3.(a) shows the resistivity is lower with low target substrate distance and gas perssure condition, because of the substitution of Oxygen vacancy by Oxygen ion in native defect.[2]

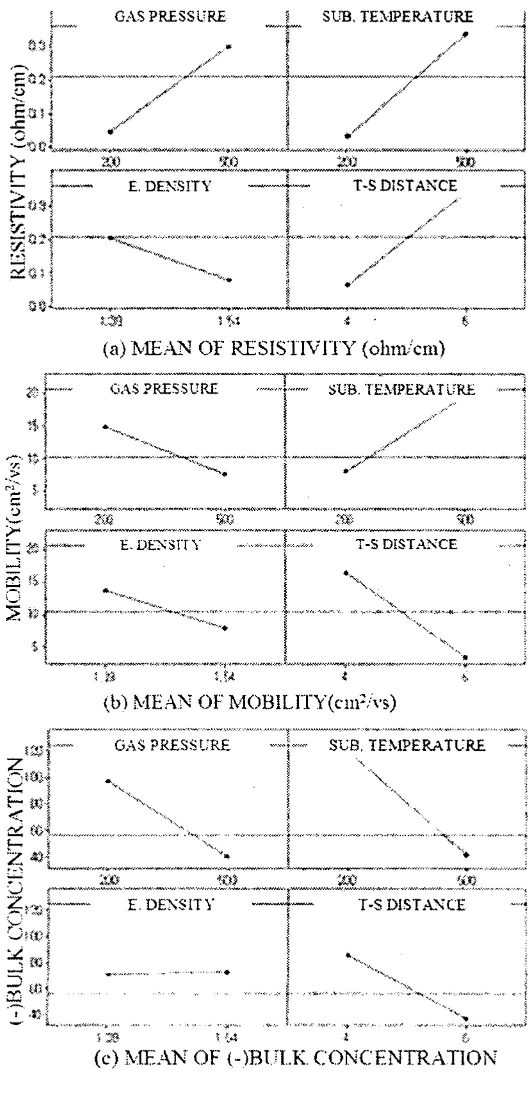

Fig 2. (a)Main effects plot(data means) for resistivity(ohm/cm) (b)Main effects plot (data means) for mobility(cm2/cm) (c)Main effects plot for (-)bulk concentration

In Fig. 3(b) shows the mobility is higher with low T-S distance and high energy density condition. In Fig. 3(c) shows the bulk concentration is higher with low gas pressure and target substrate distance.

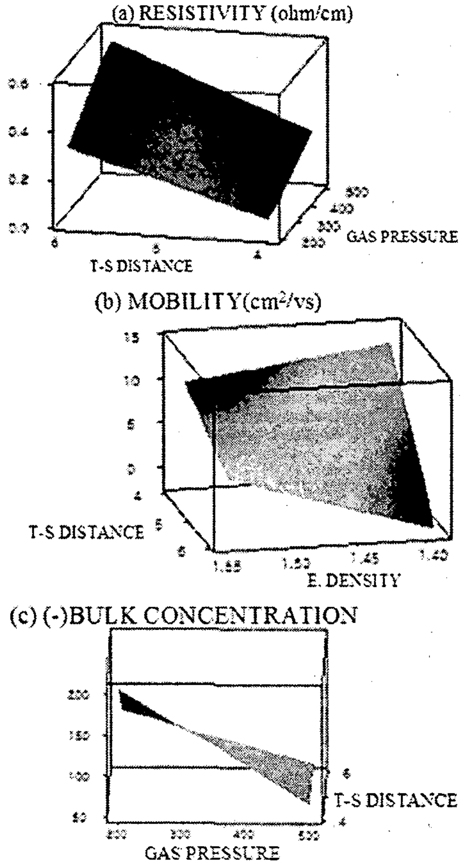

Fig 3. (a) Surface plot of resistivity of T-S distance vs Gas pressure (b) Surface plot of mobility of T-S distance vs Energy density (c) Surface plot of concentration of T-S distance vs Gas pressure

4. Conclusion

It has been investigated that Ga-doped ZnO(GZO) films deposited on a sapphire utilizing pulsed laser deposition method using ZnO and Ga_2O_3 targets. As-grown GZO microstructure and electrical properties try out using by XRD and hall measurement. The interactions of input factors were analyzed by using DOE. According to the results of our experiments, resistivity becomes lower with low target-substrate distance and gas pressure condition, and mobility becomes higher with low distance and high energy density target substrate distance. The bulk concentration becomes higher with low gas pressure and low target substrate distance.

References

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