

# EFFECT OF OXYGEN PRESSURE ON THE CHARGE TRANSPORT PROPERTY OF AL-DOPED ZnO FILMS GROWN BY PULSED ELECTRON DEPOSITION

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**Abstract.** *Al-doped ZnO films were grown by pulsed electron deposition (PED) in an oxygen pressure ranging from 0.4 Pa to 2.7 Pa at room temperature and at 400 °C. Charge transport properties were measured by Van der Pauw technique. Crystallinity of the films was characterized by X-ray diffraction. It was found that the films grown at room temperature have a very high resistivity due to poor crystallinity. For the films grown at 400 °C, the obtained results indicate that the film grown at a lower pressure has a higher carrier mobility as well as a higher carrier concentration, resulting in a lower resistivity. This phenomenon has been discussed in term of the concentration of oxygen vacancies and the number of traps for carriers.*

## I. INTRODUCTION

Recently, much attention has been paid on zinc oxide which is highly transparent semiconductor in the visible range of wavelengths with a wide band gap of about 3.37 eV at room temperature and a high excitonic binding energy of about 60 MeV [1]. When doped with group-III elements like Al, Ga and In, its resistivity could be reduced to  $2-4 \times 10^{-6} \Omega \times m$  [2-5]. With these features, ZnO is a promising material for fabricating a variety of devices such as transparent conducting electrode for flat panel displays and solar cells, grating in optoelectronic devices, windows in antireflection coating and optical filters [6-11]. There are a wide variety of methods for producing ZnO for fundamental studies or fabrication of devices [12-18]. Recently, pulsed electron deposition (PED) have been considered as a good method for the fabrication of high-quality ZnO thin films. In the PED technique instead of photons, energetic electrons are used to ablate targets. Therefore, this method will work with materials that are transparent. Besides the advantage as in pulsed laser deposition (PLD) that the stoichiometry of target materials is preserved in the film, PED are scalable, simple, and low cost, making this technique become a suitable tool for growing films of complex materials.

Growth of pure ZnO films by PED has been reported by Porter et al. [19], Nistor et al. [20] and Zhan et al. [21]. In previous work [22] we have studied the effect of substrate temperature on the Al-doped ZnO films deposited at 1.3 Pa. The obtained films exhibited a high transmittance but a low electrical conductivity. The lowest resistivity of  $3.4 \times 10^{-4} \Omega \times m$  has been obtained for the films grown at 400 °C. This value is still large

and not suitable for applications. We suggested that the conducting feature of these films mainly relates to the number of Al atoms really activated in matrix oxide to produce extra electrons in the band. In this work we studied the effect of oxygen pressure on the charge transport property of Al-doped ZnO films. We expected that at a lower oxygen pressure during deposition a larger number of oxygen vacancies may be generated, resulting in the increase of conductivity.

## II. EXPERIMENT

Al-doped ZnO (AZO) thin films were deposited on glass and quartz substrates using PED technique. The pulsed electron gun used in our experiments is a commercial source, PEBS-20 manufactured by Neocera, Inc. The technical specification of the PED system has been described elsewhere [22]. For all films, the discharge voltage was maintained at 14 kV and the pulse frequency was maintained at 5 Hz. A discharge voltage of 14 kV was chosen, this is because, according to a recent report of Strikovski et al. [23], this is the optimal potential for obtaining a maximum deposition rate. The films were grown at both the room temperature and 400°C under an oxygen pressure ranging from 0.4 Pa to 2.7 Pa. The oxygen pressure of 0.4 Pa is the lowest limit pressure for the PED system used in our work. The target used was a high purity 2 wt % Al<sub>2</sub>O<sub>3</sub>doped ZnO target of diameter 2 inch and thickness 3 mm. All samples were deposited with 20,000 electrical pulses.

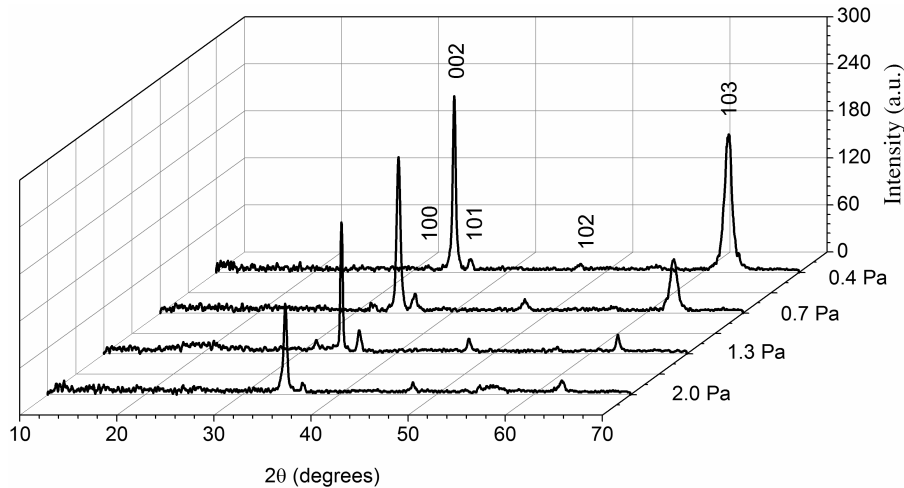
The crystallinity of the films was characterized by X-ray diffraction using Cu-K<sub>α</sub> radiation (XRD, Bruker D5005) in a special configuration where the incident angle was kept at 1.5°. Thickness was examined by stylus profiler (Veeco- Dektark D150). The transport properties was measured by Van der Pauw technique at room temperature (Quantum Design- Ever Cooler).

## III. RESULTS AND DISCUSSION

The thickness of the films determined by stylus technique decreased with the increase of the oxygen pressure, as seen in Table 1. This variation of the thickness can be explained due to the interaction between incoming ions in plasma flux and gas atoms, resulting in a decrease of current of ions arriving at the substrate.

The effect of oxygen pressure on the crystallinity of the films grown at room temperature has been reported in the previous work [22], where the authors indicated a common situation of the poor crystallinity. However, among these films, the ones grown at lower oxygen pressure (i.e. 0.4, 0.7 and 1.3 Pa) more or less exhibited a wurzite structure with a preference for the [002] orientation whereas the others grown at higher oxygen pressure (i.e. 2.0, 2.7 Pa) exhibited amorphous structure.

Figure 1 shows the XRD patterns of the films grown at 400°C and at different oxygen pressures. From this figure one can see that all the films exhibited a clear wurzite structure with a highly preference for the [002] orientation. In addition, the crystallinity evaluated from the intensity and a full width at half-maximum (FWHM) of the [002] peak improves the decrease of the oxygen pressure. This phenomenon may relate to the fact that the value of the oxygen pressure is optimal one originating from the requirement of the average energy per deposited atom which should be about 10-20 eV. This energy, in one hand,



**Fig. 1.** XRD patterns of the films grown at 400 °C and at different oxygen pressures.

is strong enough to complete the disruption of the columnar morphology of the growing film, but in other hand, it does not damage the surface. It is well-known that in both the PED and (PLD) technique, with the presence of background gases during ablation, the species arriving at substrate lose their average velocity as they undergo through scattering, thermalization and deceleration.

Electrical resistivity ( $\rho$ ) have been calculated from the measured four-probe resistance ( $R$ ), using the relation  $\rho = (w.t/l)R$ , where  $w$  is the width,  $t$  is thickness and  $l$  is the length of the sample. The electrical resistivity of the films grown at room temperature and at different oxygen pressures is listed in Table 1. The Hall measurement for these films was not appropriate because of their high resistivity. However, it is possible to realize that the resistivity is lower for the films deposited at lower oxygen pressure. In these films, where the crystallinity is poor, most carriers are oxygen vacancies. This fact explained the decrease of resistivity with decreasing oxygen pressure. A very high resistivity of the films originated from both the large number of defects acted as the traps for carriers and the low carrier mobility.

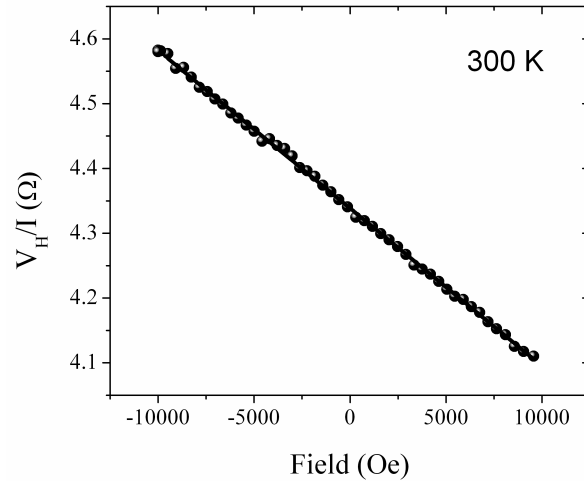
**Table 1.** The thickness and electrical resistivity of the films grown at room temperature as a function of oxygen pressure

Oxygen pressure (Pa)	Thickness (nm)	Substrate temperature (°C)	Resistivity ( $\Omega \times m$ )
0.4	280	25	$7 \times 10^{-4}$
0.7	270	25	$3 \times 10^{-3}$
1.3	250	25	$5 \times 10^{-2}$
2.0	230	25	$6 \times 10^{-1}$
2.7	220	25	$4 \times 10^0$

The resistivity vs. oxygen pressure for the films grown at 400°C is listed in Table 2. One can see that the resistivity of these films is much lower than that of the films grown at room temperature due to a better crystallinity. This phenomenon can be explained due to the growth of crystallinities, leading to the increase of the number of Al atoms really activated in matrix to produce extra electrons in the band as well as to the decrease of number of traps. The most interesting behavior is the oxygen pressure dependence of resistivity, i.e. the resistivity decreases dramatically as the oxygen pressure decreases and reaches to a value  $2.4 \times 10^{-5} \Omega \times \text{m}$ . This value is one order higher than that obtained by Anders et al. [24] where the pulsed filtered cathodic technique was used (about  $5 \times 10^{-6} \Omega \times \text{m}$ ) and by Dikovska [4] where the PLD technique was used ( $2.4 \times 10^{-6} \Omega \times \text{m}$ ).

**Table 2.** The electrical resistivity, carrier mobility and carrier concentration of the films grown at 400°C as a function of oxygen pressure.

Oxygen pressure (Pa)	Resistivity ( $\Omega \times \text{m}$ )	Carrier mobility ( $\text{m}^2/\text{Vs}$ )	Carrier concentration ( $\text{m}^{-3}$ )
0.4	$2.4 \times 10^{-5}$	$7.2 \times 10^{-4}$	$3.7 \times 10^{14}$
0.7	$5.8 \times 10^{-5}$	$5.9 \times 10^{-4}$	$1.8 \times 10^{14}$
1.3	$3.2 \times 10^{-4}$	$3.8 \times 10^{-4}$	$5.1 \times 10^{13}$
2.0	$7.6 \times 10^{-4}$	$2.2 \times 10^{-4}$	$3.7 \times 10^{13}$
2.7	$10^3$	x	x



**Fig. 2.** A typical result of Hall measurement carried out on the film grown at 400°C and at the oxygen pressure of 3 mTorr.

The behavior observed for resistivity of the films grown at 400°C can be explained due to both the carrier concentration and the carrier mobility that was determined from the Hall measurements. In our Hall measurements, the ratio  $V_H/I$ , where  $V_H$  is the Hall voltage,  $I$  is the current through the sample, was measured as a function of the applied

magnetic field ( $H$ ) normal to the sample plane. Figure 2 shows a typical result of the Hall measurement. From the equation  $V_H = I H / ent$ , where  $e$  is the electron charge and  $n$  is carrier concentration, one can expect that  $V_H / I = H / ent$  is a linear function with a slope  $a = 1 / ent$ . The negative sign of the slope indicates that the majority carriers are electrons. Carrier concentration ( $n$ ) has been determined from equation  $n = 1 / (aet)$ . The carrier mobility has been extracted using the relation  $\mu = 1 / (en\rho)$ . The carrier concentration and carrier mobility are also listed in Table 2. Again, it was impossible to measure Hall effect for the films grown 2.7 Pa because of their high resistivity. However, one can see that the increase of both concentration and carrier mobility with decreasing oxygen pressure leads to an antagonistic variation of resistivity. Concerning to the variation of carrier concentration with oxygen pressure, one can suggest that the lower oxygen pressure would produce more oxygen vacancies whereas the higher oxygen pressure would bring about more absorptions of oxygen in the grain boundaries which act as traps for the carriers. This suggestion is consistent with the proposal of Nunes et al. [25] where the authors explained the effect of annealing process under the oxidation and reduction atmosphere on the carrier concentration of ZnO films. Because the carrier mobility relates to the scattering phenomena, the dependence of carrier mobility on oxygen pressure can be explained due to the improvement in crystallinities as decreasing oxygen pressure.

#### IV. CONCLUSION

It has been observed that the films grown at lower pressure has a higher carrier mobility as well as a higher carrier concentration, leading to larger conductivity. This improvement in electrical conductance has been explained due to the increase of concentration of oxygen vacancies and the decrease of number of traps for carriers. The obtained results show possibilities to reduce the resistivity of ZnO films not only by substitutional doping higher valence elements into Zn atoms, but also by non-stoichiometry of the films resulting from oxygen vacancies.

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