Study the Effect of ZnO/Cu/ZnO Multilayer Structure by Radio Frequency Magnetron Sputtering for Flexible Display Applications

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Abstract

ZnO/Cu/ZnO multilayer structured glass and Polyethylene naphthalate (PEN) with different thicknesses of copper (Cu) layer were prepared by RF magnetron sputtering at room temperature. The electrical resistivity of the films was achieved at $8.6 \times 10^{-5} \Omega \cdot \text{cm}$ and $4.5 \times 10^{-4} \Omega \cdot \text{cm}$, the carrier concentration of $5.32 \times 10^{21} \text{ cm}^{-3}$ and $1.36 \times 10^{21} \text{ cm}^{-3}$, low sheet resistance of $12 \Omega/\text{cm}^2$ and $16 \Omega/\text{cm}^2$ respectively on glass and PEN substrate respectively with the optical transmission of more than 80% for both cases at the optimum copper layer thickness. The conduction mechanism of the multilayer structure has been explained in terms of metal-to-oxide carrier injection with the variation of the thickness of the Cu layer. Different optimization procedures were used to achieve good optically transparent and electrically conductive films. The structural characterizations of multilayer films were carried out by X-ray diffraction (XRD); surface morphology and topography were performed by scanning electron microscopy (SEM) and atomic force microscopy (AFM) respectively. The value of the sheet resistance of the structure was influenced by the deposition condition of both layers. The effect of the substrate and thickness of the Cu layer was investigated and illustrated.

Keywords: RF magnetron sputtering; Flexible Display Applications; ZnO/Cu/ZnO Multilayer Structure.

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1. Introduction

Transparent and conducting oxide (TCO) is a type of material that can show conductivity and transparency simultaneously.^[1-2] TCO films show excellent electrical resistivity in the order of $10^{-4} \Omega \cdot \text{cm}$ to $10^{-5} \Omega \cdot \text{cm}$ with an optical transmission of more than 90%. The balanced electrical and optical properties of TCO makes it useful for a wide range of applications in flat-panel displays, solar cells, gas sensors, thin film transistor (TFT), liquid crystal displays (LCD), photovoltaic devices, organic light emitting diodes (OLEDs) and smart windows.^[3-8] TCO films can be deposited by different deposition techniques such as spray pyrolysis,^[9] sol-gel process,^[10] pulsed laser deposition,^[11] thermal evaporation,^[12] chemical vapor deposition,^[13] electron beam deposition^[14] and radio frequency (RF) magnetron sputtering^[15-17] on various substrate. Among different deposition techniques RF magnetron sputtering

technique is more beneficial due to its uniformity and high deposition rates.^[18, 19]

Indium tin oxide (ITO) and Fluorine doped tin oxide (FTO) are commercially established as highly efficient TCO materials. These materials show excellent electrical conductivity with the high optical transmission in the visible region of the solar spectrum. The cost and availability of indium materials influenced the thinking of researchers to find out alternative TCO materials. Different doped (Al, Ga, In) zinc oxide (ZnO) films exhibit good electrical conduction and optical transmission in the visible region.^[20-21] In different experiments many groups have shown promising results of electrical resistivity ($10^{-4} \Omega \cdot \text{cm}$) and high optical transmission (more than 90%) in the case of doped Zinc oxide (ZnO)^[22,23] thin films. The results such as conductivity, optical transmission, and sheet resistance of doped ZnO thin film, make it an alternative TCO material. Such types of properties make ZnO films appropriate materials for optoelectronic devices, solar cells, and OLEDs. ZnO materials have some advantages such as easy availability, low cost, good radiation resistance, good conductivity, and high optical transmission.^[24] Only there is a limitation for increasing the conductivity, as it

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is known that the conductivity can increase with increasing the carrier concentration, due to its ionized impurity scattering. It is found that synthesized ZnO thin films at a temperature from 200 to 300 °C are polycrystalline and textured. ZnO thin films, which are synthesized using pure Ar plasma gas, are highly porous and show very good optical transmittance and absorption spectra (near 380 nm).^[25] Abed *et al.* studied the effect of RF magnetron on conventional solid-state targets based on ZnO-MgO: Al₂O₃ (10:2 wt %) material on glass and silicon substrates at different working pressures. The resistivity of $8.8 \times 10^{-2} \Omega\text{-cm}$ was achieved by the sample deposited at the lowest working pressure of 0.21 Pa.^[26] Recently, the annealing effect on the structural and optical behavior of (ZnO: Eu³⁺) thin film growth has been studied by Otieno *et al.*^[27] In our recent publication, we studied the effect of substrate and substrate temperature on the microstructure of magnetron sputtering doped-ZnO thin films.^[28] For a few applications where low sheet resistance and high optical transparency are required still not achieved. Different research groups had reported the dielectric-metal-dielectric thin film with different metals such as silver, Ag, and Cu with increasing conductivity and high optical transmission.^[29-31]

Among them, the introduction of the Cu layer in a multilayer structure plays a significant role in increasing the conductivity of the thin film. But the reports on the effect of thin Cu layer in multilayer structures are very few. To improve the overall performance of the transparent conductive films, Rasheed *et al.*,^[32] Wang *et al.*^[33] and Toma *et al.*^[34] studied structural, electrical, and optical properties of ZnO/Cu/ZnO multilayer film stacking using Cu film as a sandwich layer. The results showed that ZnO/Cu/ZnO multilayer film has good crystalline properties and with an increasing Cu layer thickness, visible light transmittance of the multilayer film is reduced, while the electrical performance improved significantly. Sahu *et al.* studied the influence of post-growth annealing on the structural, electrical, and optical properties of the multilayer ZnO/Cu/ZnO films, prepared on glass substrates by simultaneous RF magnetron sputtering of ZnO and dc magnetron sputtering of Cu.^[35] Recently scientists are focusing their work to develop and replace the glass substrate with a flexible substrate for application in the display, optoelectronics, and mobile. Lightweight, thermally stable, chemically resistant, and dimensionally stable properties are preferred for a flexible substrate. Among the different

Fig. 1 Variation of (a) sheet resistance, (b) mobility, (c) figure of Merit, and (d) carrier concentration of ZnO/Cu/ZnO with a variation of different Cu thickness.

alternative flexible substrate polyethylene naphthalate (PEN) was one of them studied recent past by different research groups. In this paper, we have studied the optical, and electrical properties and figure of merit of ZnO/Cu/ZnO multilayer structure by optimizing ZnO and Cu thickness at room temperature on glass and polyethylene naphthalate (PEN) substrate.

2. Experimental

Coating of ZnO and Cu multilayer thin films was performed on glass and PEN substrates by dual-target RF magnetron (powered at 13.56 MHz radio frequency) sputtering system room temperature (RT) under the nonreactive environment with RF power 100W. ZnO and Cu discs with a purity of (99.99 %) having a 2-inch diameter and 5 mm thickness were used as a sputtering targets. The process chamber is evacuated up to a base vacuum of 2.4×10^{-6} torr. Target to substrate distance was kept at 6 cm for all experiments. Argon was used as inert gas ambient and constant at 40 sccm throughout the entire deposition process. Before final deposition pre-sputtering of the ZnO target is done in a pure argon plasma atmosphere for about 15 min in order to remove the surface oxide layer of the target. And before that, the glass substrate was cleaned properly with soap, methanol, and an ultrasonic bath.

The thickness of the film was measured using a digital thickness monitor (DTM) attached to an RF magnetron sputtering system. The electrical properties of the multilayer (ZnO/Cu/ZnO) films were studied by the Ecopia-HMS-3000 set-up. Optical transmittance and absorbance data of multilayer thin films were measured using a UV-VIS spectrophotometer. Structural characterization of the films was carried out by X-ray diffraction (XRD) (Philips PW 1710 diffractometer) (Cu K α , $k = 1.54178$ Å, 2θ scan mode). The surface morphology and topography were performed by Atomic Force Microscopy (AFM) (Tap 300 G).

3. Result and Discussion:

ZnO/Cu/ZnO films show promising electrical properties with $\rho = 8.6 \times 10^{-5} \Omega \cdot \text{cm}$, $R_{\text{sh}} = 12 \Omega/\text{cm}^2$ and $\rho = 4.5 \times 10^{-4} \Omega \cdot \text{cm}$ with $R_{\text{sh}} = 16 \Omega/\text{cm}^2$ and optical transmission of 86% and 84%, respectively on glass and PEN substrate. The resistivity, mobility, and carrier concentration have been estimated from Hall measurements. The dependence of the resistivity, Hall mobility, and carrier concentration of ZnO/Cu/ZnO multilayer with a variation of Cu layer thickness are shown in Fig. 1. It shows (Fig. 1) the sheet resistance (R_s) as a function of the Cu layer thickness. Sheet resistance was decreased with increases in Cu layer thickness. A similar kind of result has been reported by Luan *et al* in another study.^[36] With increasing the Cu thickness the surface of the Cu thin films shows shifted from an island state to a continuous state. After the formation of the continuous film sheet resistance begins to decrease. The mobility factor doesn't have any effect on the sheet resistance. The mobility of multilayer film with different thicknesses of

the copper layer is shown in Fig. 1. Mobility of the multilayer was increased with the thickness of the Cu layer.

The optical transmittance of the multilayer (ZnO/Cu/ZnO) thin films on Glass and PEN substrates are shown in Fig. 2. The thin films of multilayer show the highest transmittance 86% and 84% on glass and PEN substrates respectively, in the visible region of the solar spectrum. As the thickness of the Cu layer increases from 2 nm to 8 nm optical transmittance decreases from 85% to 80% and 82% to 78% on glass and PEN substrates respectively. The decrease of transmittance in the higher region of the solar spectrum (near-infrared) results from the increase of reflectance and absorption which is attributed to the plasma resonance of the electron gas in the conduction band. The quality of good transparent conducting films can be judged by the figure of merit (FOM). The figure of merit was calculated by the equation $\phi = T^{10}/R_s$ where (T) transmittance and (R_s) sheet resistance.^[37] In the case of glass and PEN substrate multilayer film (with 4nm Cu layer) shown (Fig. 1) highest FOM is $1.84 \times 10^{18} (\Omega/\text{cm}^2)^{-1}$ and $1.09 \times 10^{18} (\Omega/\text{cm}^2)^{-1}$, respectively. A higher value of the figure of merit means better performance of transparent conducting oxide film. The electrical and optical properties of multilayers are mainly dependent on Cu properties.

Fig. 2 Optical Transmission of ZnO(50nm)/Cu(4nm)/ZnO(50nm) multilayer on Glass (1) and PEN substrate (2).

Structural analysis of Cu layer-based multilayer structure was investigated by XRD. XRD pattern of room temperature deposited multilayer on glass and PEN substrate was shown in Fig. 3. Films are showing a strong ZnO (002) peak at $2\theta = 34.33^\circ$ representing the hexagonal wurtzite polycrystalline structure with strongly preferred c-axis orientation. Variation of Cu layer thickness from 2nm to 8 nm, the intensity and ZnO peak do not change. It is also clear from the XRD that a small peak has been observed Cu (111) peak at $2\theta = 43.21^\circ$. As the thickness of the Cu layer increases peak gradually becomes stronger with FWHM (Full Width Half Maximum) peak

Fig. 3 XRD patterns of ZnO/Cu/ZnO multilayer structure with the different thickness of Cu layer on Glass and PEN substrate.

gradually decreases. This is a sharp indication of improved crystallinity with the increase of the Cu thickness. Optimum electrical and optical properties were achieved for a multilayer with a 4nm Cu layer.

The optical band gap of the ZnO/Cu/ZnO multilayer on glass and PEN substrate is shown in Table 1. Variation of band gap was shown with different thicknesses of the Cu layer. Optical bandgap was calculated by using the equation $\alpha h\nu \propto (h\nu - E_g)$ where α stands for the absorption coefficient, $h\nu$ optical energy, and E_g represents the optical band gap.^[38] It is clear from the table that the optical bandgap increases with decreasing Cu thickness. Decreasing bandgap may be because some Cu atoms permeate ZnO at the interface of the ZnO/Cu/ZnO multilayer. The value of bandgap with different thicknesses shows lower than the bulk of ZnO (3.37 eV).

Table 1. The band gap of a multilayer with different thicknesses of Cu is on Glass and PEN substrate.

Substrate	ZnO/Cu/ZnO	Bandgap (eV)
Glass	ZnO(50nm)/Cu(2nm)/ZnO(50nm)	3.20
	ZnO(50nm)/Cu(4nm)/ZnO(50nm)	3.15
	ZnO(50nm)/Cu(6nm)/ZnO(50nm)	3.14
	ZnO(50nm)/Cu(8nm)/ZnO(50nm)	3.12
PEN	ZnO(50nm)/Cu(2nm)/ZnO(50nm)	3.18
	ZnO(50nm)/Cu(4nm)/ZnO(50nm)	3.16
	ZnO(50nm)/Cu(6nm)/ZnO(50nm)	3.14
	ZnO(50nm)/Cu(8nm)/ZnO(50nm)	3.11

ZnO and Cu have a work function of 5.4 eV and 4.5 eV respectively.^[39] It is clear from the figure that the work function of copper is lower than the ZnO film and an accumulation layer form at the interface due to the transfer of electrons from the metal to the semiconductor. The energy band diagram of the Cu/ZnO structure under isolation and interface is shown in Figs. 4(a) and 4(b), respectively. The interface of Cu and ZnO (Fermi level) aligns in the interface region due to the transfer of electrons from copper to ZnO. The transfer of electrons makes the valence and conduction bands of ZnO bend downward [Fig. 4(b)]. Work function differences

between Cu and ZnO are large as a result significance of carriers in the ZnO semiconductor layer.

Fig. 4 Schematic energy band diagrams of Cu and ZnO (a) before contact and (b) after contact.

It is clearly seen that the substantial decreases in resistivity and sheet resistance of the multilayer structure. Fig. 1 shows that Cu thickness, the ZnO/Cu/ZnO multilayer structure has a lower Hall mobility at 4 nm Cu layer in both glass and PEN substrate. This result suggests that most of the current pass through the ZnO layer with a thin Cu layer which acts as discontinuous scattering sites that reduce the mobility. It is clearly seen from the Fig. 1 that as the thickness of the Cu layer increases, the carrier concentration increases from $8.68 \times 10^{20} \text{ cm}^{-3}$ to $7.68 \times 10^{22} \text{ cm}^{-3}$ and $7.20 \times 10^{20} \text{ cm}^{-3}$ to $8.21 \times 10^{20} \text{ cm}^{-3}$ respectively for glass and PEN substrate. Also, the mobility increases from $16.24 \text{ cm}^2/\text{Vs}$ to $17.02 \text{ cm}^2/\text{Vs}$ for the glass substrate. These results indicate that the Cu layer becomes near continuous to electron conduction and a large number of current passes through it. Basically, the sandwich layer of two ZnO and a thin Cu layer is a parallel network of electrical resistors. The amount of current flowing through each of the individual layers in turn is inverse to their resistance. Resistivity of the multilayer reduces from $4.2 \times 10^{-4} \Omega \cdot \text{cm}$ to $8.6 \times 10^{-5} \Omega \cdot \text{cm}$, $5.0 \times 10^{-4} \Omega \cdot \text{cm}$ to $4.5 \times 10^{-4} \Omega \cdot \text{cm}$ for

Fig. 5 AFM images of ZnO/Cu/ZnO thin film on a) Glass (Cu 2nm) b) Glass (Cu 4nm) and c).PEN substrate (4nm).

glass and PEN substrate respectively with the increasing of the thickness

of the Cu layer. A multilayer of ZnO/Cu/ZnO with 4nm Cu thickness shows a better percolation threshold.

The surface topography of ZnO/Cu/ZnO thin films grown on glass and PEN substrate is shown in the AFM image (Fig. 5). Fig. 5(a) and Fig. 5(b) show the AFM image of ZnO/Cu/ZnO on a glass substrate with Cu thickness of 2 nm and 4 nm respectively. From this figure, it is clear that Fig. 4(a) shows non-uniform of crystallites with an amorphous phase whereas Fig. 5(b) shows uniform spherical grain distribution. Also, the surface looks very smooth these properties are appropriate for display applications. The surface morphology of Multilayer films with Cu 4 nm deposited on PEN substrate is shown in Fig. 5(c). The surface on the PEN substrate also looks smooth which was appropriate for display applications.

Table 2. Surface roughness, the grain size of ZnO/Cu/ZnO multilayer with different thicknesses of Cu films.

Substrate Type	ZnO(50nm)/Cu/ZnO(50nm) Variation of Cu (nm)	Surface roughness (δ_{rms} in nm)	Grain size (nm)
Glass	2	8.64	19
	4	8.32	18
	6	7.42	16
	8	7.12	14
PEN	2	9.89	24
	4	8.75	17
	6	8.45	14
	8	7.68	2

Variations of grain size and surface roughness with different thicknesses of Cu in Glass and PEN substrates are shown in Table 2. Among other properties, surface roughness is one of the important properties for many optoelectronic and flexible display applications. A smooth surface is appropriate for OLED and display applications whereas a rough surface is preferable for solar cells. A rough surface increases the scattering of the light as a result optical transmission increases and finally, it enhances the efficiency of the Cell. [40-45]

4. Conclusion

Multilayer films of ZnO/Cu/ZnO structure show high transmission in the visible range of the solar spectrum with a low sheet resistance of $12 \Omega/\text{cm}^2$ by RF magnetron sputtering techniques. The good electrical resistivity ($8.6 \times 10^{-5} \Omega \cdot \text{cm}$) and optical transmission (86%) of the multilayer films on glass are achieved by the optimization of growth condition. Also, the 4 nm thick copper layer in the structure shows carrier concentrations $5.32 \times 10^{21} \text{ cm}^{-3}$ and $1.36 \times 10^{21} \text{ cm}^{-3}$, with the figure of merit 1.84×10^{18} and 1.09×10^{18} glass and PEN substrate respectively. Electrical conduction and optical transmission were interdependent properties among others mainly affected by film structure. Improved electrical and optical properties also depend on preparation process parameters. Due to this advantage, RF magnetron sputtering is found to be the most favorable deposition method. The properties such as electrical and optical of the multilayer films depended dominantly on the characteristics of the metal interlayer. To achieve low resistivity and high optical transmission, the optimum thickness of the copper layer plays a crucial role with minimal loss in transmission due to absorption, reflection, and scattering. The morphology and structure of the very thin metal layer were also sensitive to the deposition condition. Room temperature deposited films shows excellent conductivity and optical transmission compared to those commercial indium tin oxides. The result makes the ZnO/Cu/ZnO multilayer an appropriate candidate for optoelectronic devices and flexible display applications.

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Conflict of interest

There are no conflicts to declare.

Supporting information

Not applicable

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