Surface characterization of ZnO:Al transparent thin films

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Abstract— *We present the effect of hydrogen on surface characterization by Rf magnetron sputtered ZnO:Al thin films. ZnO:Al films are self textured and surface roughness of ZnO:Al films varies with H_2 dilution ratio and ultimately affects on diffused transmittance varies from 41-27% with the variation of hydrogen (5-40%) with improved resistivity of 3.9×10^{-4} Ω-cm. The light scattering effect of hydrogen textured thin films was evaluated by the measurement of diffused transmittance. Experimental result shows that introduction of hydrogen effect significantly improved the light trapping in terms of diffuse transmittance without affecting electrical and other optical properties of ZnO:Al thin film.*

Keywords— ZnO:Al, RF magnetron sputtering, transparent conductive oxides, Atomic force microscope, Diffused Transmittance.

I. INTRODUCTION

Transparent conductive oxides (TCOs) have been extensively studied because they are essential elements in thin film optoelectronic devices applications such as thin film solar cells, flat-panel displays, and light emitting diodes [1-3].Transparent conducting oxide (TCO) thin film is a special type of materials with wide band gap oxide (>3eV), high optical transmittance (≥ 85%) over wide range of solar spectrum, and low sheet resistance. Main important TCOs are Fluorine doped tin oxide (FTO), Indium tin oxide (ITO), Aluminum doped zinc oxide (ZnO:Al), Antimony doped tin oxide (ATO), Galium doped Zinc Oxide (ZnO:Ga), Galium doped Indium Zinc Oxide (IZO:Ga), and Indium doped Cadmium Oxide thin films. Other Ternary Compounds based TCO Materials are Zn_2SnO_4, $MgIn_2O_4$, $CdSb_2O_6$:Y, $ZnSnO_3$, GaInO, $Zn_2In_2O_5$ and $In4Sn_3O_{12}$ etc. Among all TCO thin films Fluorine doped tin oxide (FTO), and Indium tin oxide (ITO) are commercially available for different device applications and the others are under lab scale development stage. TCO thin films have wide applications such as in micro-electronic devices, displays, thin film transistor, light emitting diodes (LEDs), solar cells and other photonic devices [4-9]. Though indium tin oxide (ITO) film is extensively applied to photovoltaic devices and flat panel display because of its good electrical and optical properties, it has some problems such as high cost, low stability to H2 plasma and toxicity. Aluminum-doped zinc oxide thin films are make a compete as transparent conductive oxide (TCO) films prepared materials due to the advantages of ZnO:Al films are cheap and abundant elements [10]. In addition, AZO thin films have an excellent chemical stability and specific electronic/optical properties of a wide band gap (Eg=3.4 to 3.9) semiconductor. Therefore, AZO thin films are usually used as transparent conducting electrodes in solar cells. Several studies using different deposition methods have been reported, such as sol-gel processes [11], pulsed laser deposition [12], sputtering [13] and molecular beam epitaxy [14]. RF (Radio-Frequency) Sputtering method is an effective technique due to its ability to produce reasonable quality thin films at a high deposition rate [15]. Based on the progress in the previous works, it is important to better understand the influences of rf magnetron sputtering at different Ar flows and sputtering power to obtain the optimum conductivity and transmittance.

In this study ZnO: Al thin films were prepared by using the rf magnetron sputtering at different hydrogen dilution with sputtering power 150W to examine the optical and electronic properties. Transmittance of all films is about 80 - 90 % in the visible range. The lowest resistivity of 3.9×10^{-4} Ω-cm (18.5Ω/sq) and highest transmittance of 90 % was obtained at sputtering power of 150 W and Ar flow of 40 sccm. The observed property of the ZnO:Al thin films is suitable for transparent conductive electrode applications.

II. EXPERMENTS

The ZnO:Al thin films were prepared using high purity (99.99%) sintered ceramic ZnO:Al (ZnO:Al 98:2 wt%) disc [2 inch diameter and 5mm thickness] by dual-target RF magnetron (powered at 13.56 MHz radio frequency) sputtering system (Hind High Vacuum Co. (P) Ltd) on glass substrate at substrate temperature $T_s=300^0$C under Ar+H_2 environment.

Before deposition, the process chamber is evacuated up to a base vacuum of 3.47×10^{-6} bar. The RF power was varied from 50-90W with a step increment of 20W. Target to substrate distance was kept at 6 cm for all experiments. Pre-sputtering of ZnO:Al target is done in pure argon plasma atmosphere for about ten minutes in order to remove the surface oxide layer of the target before deposition of the film and the glass substrates were also plasma cleaned. The optimum deposition conditions was 80watt rf power, 5.4×10^{-3}bar, substrate temperature 300^0C substrate temp and C_H=5%.

III. RESULTS AND DISCUSSION

The crystalline structure and preferential orientation of ZnO:Al thin films were examined by X-ray diffraction(XRD).The XRD spectrum is quite similar for all the ZnO films. A typical XRD spectrum of ZnO:Al film shown in Fig.1. ZnO:Al films shows polycrystalline nature with (002) preferred orientation along C-axis with hexagonal wurtzite structure. In case of polycrystalline hexagonal structure, the most closely packed and with the lowest free surface energy is the (002) plane, which will favor the grain growth in this direction[16].The grain size of the ZnO films can be determined from the XRD spectrum by using Scherrer equation[17].

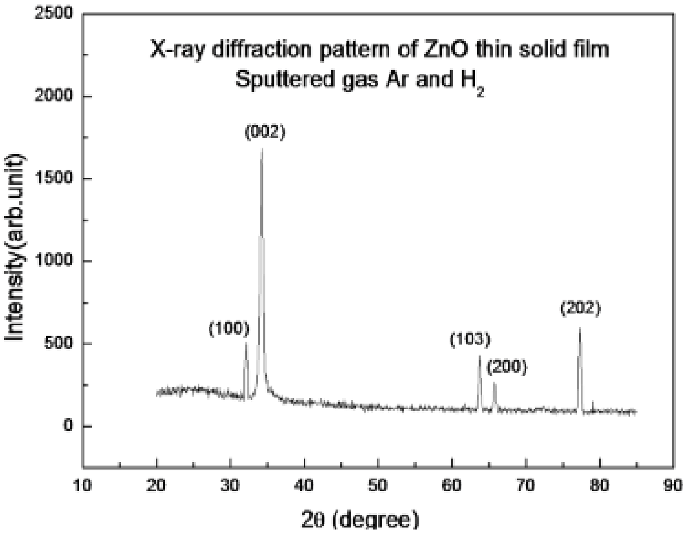

FIG.1 XRD SPECTRUM OF *ZnO:Al* THIN FILM

Table-I shows the electron concentration (n), resistivity (ρ) and mobility (μ) of ZnO:Al thin films as a function of film thickness. Lowest resistivity of 3.9×10^{-4} ohm.cm has been achieved for a value of C_H=5%

TABLE-1
ELECTRICAL PARAMETERS OF THE ZnO:Al THIN FILMS WITH DIFFERENT C_H VALUES

% C_H	Resistivity (Ω.cm)	Sheet Resistance (Ω/sq.)	Mobility cm^2/V.S	Carrier Concentration/cm^3	Grain Size	Surface Roughness
40	7.6×10^{-4}	69	6.85	1.1×10^{21}	15	9.962
20	6.2×10^{-4}	56	4.7	2.1×10^{21}	16	9.689
10	4.7×10^{-4}	42	5.8	2.3×10^{21}	18	8.876
5	3.9×10^{-4}	35.3	8.93	4.1×10^{21}	19	8.355

In this work optical property of the film was performed with the double beam spectrometer Lambda 35 from Perkin Elmer. Measurements were made using a scanning spectrophotometer fitted with an integrating sphere. By placing the test sample at the entrance of the sphere, transmittance light enters the sphere. The sphere collects all the light passing through the sample to obtain the total transmittance. Total transmittance of the ZnO:Al thin film with C_H value(1-4 with C_H value 5,10,20,40 respectively) shown in Fig.2. In case of diffused transmittance, there is a hole in the integrating sphere so that light passing directly through the sample was absorbed by a black absorber and rest of light was diffused light shown in Fig.3. This results show that the light scattering of ZnO films can be significantly improved with a optimum C_H value of 5%. Figure shows the higher diffused transmittance in case of C_H value of 5% compare to the other dilution sample of hydrogen

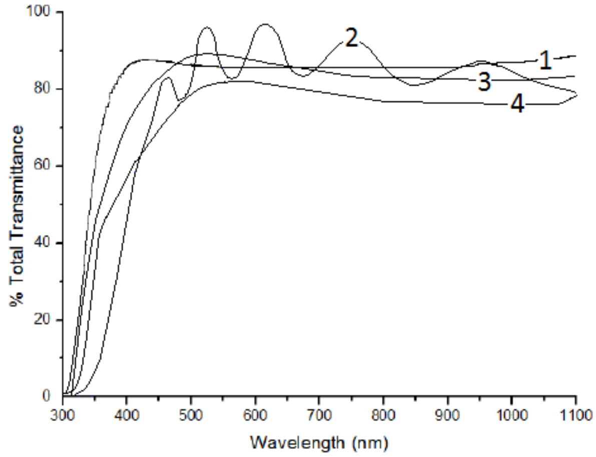

FIG.2. TOTAL TRANSMITTANCE WITH DIFFERENT VALUE OF C_H.

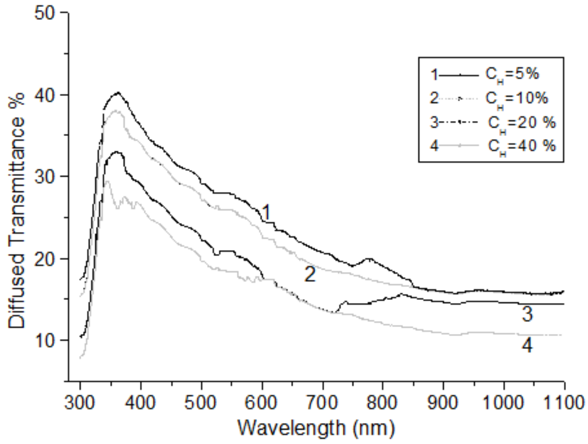

FIG.3. DIFFUSED TRANSMITTANCE WITH DIFFERENT VALUE OF C_H.

The surface topography and morphology of ZnO:Al thin films were examined using atomic force microscopy AFM. We shows 3d-view image of etched ZnO films for C_H=5%,10% & 20% respectively in fig.4.The surface roughness (rms) of ZnO films etched in hydrogen with concentration of 0%,5%,10%,20% & 40% are respectively. The surface roughness of the textured ZnO:Al thin films increases with the decreasing of hydrogen dilution.

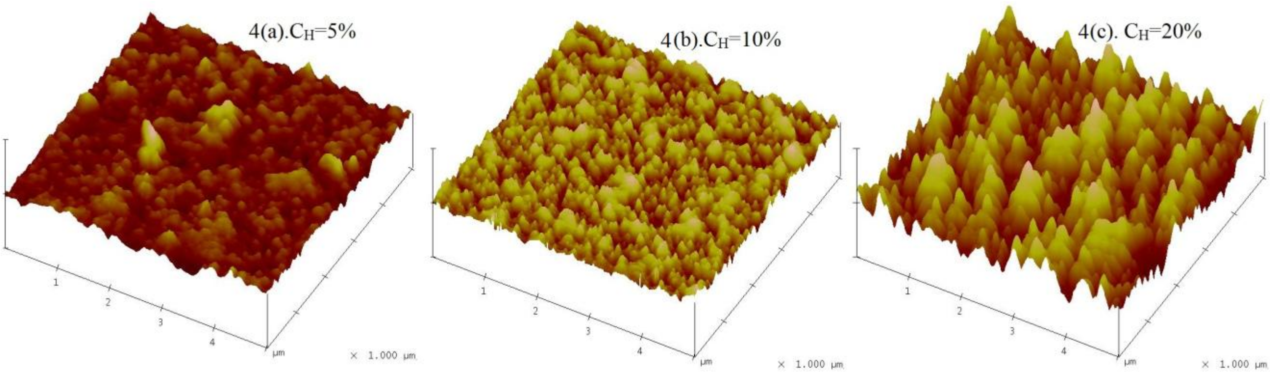

FIG.4.AFM IMAGE OF ZnO:Al THIN FILM WITH DIFFERENT VALUE OF C_H.

IV. CONCLUSION

The electrical, optical and surface morphological properties of ZnO thin films etched with hydrogen were investigated in this paper. Experimental results show that hydrogen gas incorporation with Ar plays an important role in modifying the morphology, optical and electrical properties of ZnO films. The surface roughness of the textured ZnO thin film is increasing with decreasing of the hydrogen dilution concentrating resulting in light scattering, which turn in favors light is trapping to enhance the light absorption. The diffused transmittance increases from 27% to 41% with improved low resistivity from 7.6×10^{-4} to 3.9×10^{-4} of ZnO: Al with decreasing of C_H value 40%-5%.In this study it was demonstrate that the introduced of more scattering diffused light improved the light absorption and resistivity of ZnO:Al film by proper textured surface through hydrogen gas.

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