

Growth and Characteristics of $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ Multi layer Film

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Abstract—Transparent conducting films having a hybrid structure of $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ were deposited on soda lime glass substrates at room temperature by sequential RF/DC magnetron sputtering method. The physical and optical properties of hybrid multi layered film were systematically investigated as a function of SnO_2 layer thickness. In order to estimate the optical characteristics and compare them with experimental results, the simulation program named EMP (Essential Macleod Program) was used. EMP results suggested that the multilayered thin film of SnO_2 (45 nm)/Ag (10 nm)/ Nb_2O_5 (10 nm)/ SiO_2 (10 nm)/ SnO_2 (30 nm) exhibited high transmittance of 89.7 % at 550 nm, whereas the experimentally measured transmittance showed 85.8 %, somewhat lower than simulation data. XRD patterns revealed that SnO_2 multi layered films were found to be amorphous and the surface roughness maintained a relatively small range about 4 nm.

Keywords—EMP simulation; transmittance; AES depth profiling; surface roughness

I. INTRODUCTION

Flexible transparent conductive oxides (TCOs) with low resistivity and high transmittance are of great technological importance for a wide variety application in optoelectronic, photovoltaic devices and systems such as solar cell, organic light emitting diodes, and liquid crystal displays [1-2]. So far, a number of research projects have been successfully conducted in the area of TCO materials [3-8]. The most common materials with high transmittance are SnO_2 [3], In_2O_3 [4], ZnO [5], TiO_2 [6], AZO [7], or Nb_2O_5 [8]. Recently, oxide/metal/oxide (OMO) tri-layer structures have draw great attention as TCOs with low sheet resistance, high optical transparency in the visible range and also showed relatively better chemical stability than single-layered metal film [9-15]. However, especially the superior flexibility of OMO multilayer films, which pattern can be directly exposed, poses one of the most critical issues. In order to manage visibility problem effectively, reflectance change, placing the index matching layer with high and low refractive index materials between metal and TCO, could be one of possible solutions in OMO structure. However, none of previous works have been undertaken to study the modification of optical properties through the addition of index matching layers into tri-layer structures.

In the present study, a hybrid structure of $\text{SnO}_2/\text{Ag}/\text{SnO}_2$ film with index matching layer has been designed. As an index matching material, SiO_2 ($n = 1.46$) and Nb_2O_5 ($n = 2.34$) with low and high refractive index material as well as outstanding thermal stability were selected for this purpose. The overall objective of this research is to systematically evaluate the

optical characteristics of $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SnO}_2$ multilayer film with different thickness of SnO_2 . In order to estimate the optical characteristics and compare them with experimental results in advance, the simulation program named EMP (Essential Macleod Program) was adopted.

II. EXPERIMENTAL

A. Simulation

Prior to the experiments, one of the optical programs named Essential Macleod Program (EMP) was adopted to simulate the optical characteristics such as transmittance, reflectance and color in the multi layer thin films. The EMP was processed through the following steps; first, construction parameters such as reflectance and extinction coefficient of SnO_2 , Ag, Nb_2O_5 , SiO_2 and SnO_2 , respectively, which were calculated by ellipsometry measurement, were input in the program. Second, $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ multi layers were designed and simulated with various parameters such as wavelength ranges (380 - 780 nm), thickness (10 - 50 nm) and layers of structure. Finally, analysis on the parameter effect was performed with system modification for optical properties whether it is appropriated for the optimum simulation.

B. Film Preparation

For the purpose of study, SnO_2 (40 - 55 nm)/Ag (10 nm)/ Nb_2O_5 (10 nm)/ SiO_2 (10 nm)/ SnO_2 (25 nm) multi layer film with more than about 85 % transmittance was selected after taking a number of simulation by modifying the stacking sequence and adjusting the thickness of each layer. Fig. 1 shows the schematic diagram illustrating the structure of $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ multi layer film.

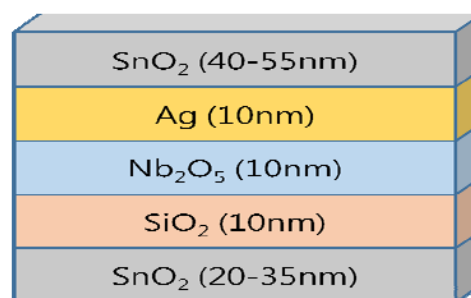


FIGURE I. THE SCHEMATIC DIAGRAM ILLUSTRATING THE STRUCTURE OF $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ MULTI LAYER FILM

A hybrid structure of $\text{SnO}_2/\text{Ag}/\text{Nb}_2\text{O}_5/\text{SiO}_2/\text{SnO}_2$ was deposited on soda lime glass substrates by sequential RF/DC magnetron sputtering at room temperature. Prior to deposition, soda-lime glass ($75 \times 25 \times 1 \text{ mm}^3$) was ultrasonically cleaned

in acetone, ethanol and IPA for 30 min at 50 °C. Thin film layers of SnO₂, SiO₂, and Nb₂O₅ were deposited by RF magnetron sputtering onto soda-lime glass substrates at room temperature. High purity Ar (35 sccm) was introduced into the chamber by mass flow meter with the total pressure maintained at 5.5 mTorr. Ag layer was deposited by DC magnetron sputtering. Total film thickness was about 100 nm. More details about sputtering conditions are given in Table. 1.

TABLE I. DEPOSITION PARAMETERS AND SPUTTERING CONDITION OF SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ Multi-Layer Film

Parameter	Condition	
	Sputtering	RF
Target	SiO ₂ , Nb ₂ O ₅ , SnO ₂	Ag
Substrate	glass	
Power	100W	10W
Base pressure	5 × 10 ⁻⁵ Torr	
Working pressure	1.1 × 10 ⁻³ Torr	
Temperature	R.T.	
Atmosphere (Ar)	35 sccm	

The refractive index n and extinction coefficient k of each monolayer were evaluated using ellipsometer (Elli-SE) in the visible range from 350 to 750 nm with a step with of 5 nm. The surface morphology of the films was investigated by Atomic Force Microscope (AFM) measurements in contact mode. The transmittance and reflectance of the films were estimated using a UV-VIS-NIR spectrophotometer (KONICA-MINOLTA CM-3600d) with a light source of D65. In addition, the interfacial properties of the SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ electrodes were analyzed using AES depth profiling.

III. RESULTS AND DISCUSSION

A. Optical Properties

To extract an accurate analysis from EMP simulation, the refractive index n and extinction coefficient k are necessary. Fig. 2 represents the measured refractive index n and extinction coefficient k for SiO₂, Nb₂O₅, SnO₂ and Ag thin film in visible range, calculated by using ellipsometry. The obtained refractive indexes of SiO₂, Nb₂O₅, SnO₂ and Ag thin films were 1.52, 2.42, 2.09 and 0.12, respectively, which are somewhat higher than theoretical value. The slightly high value of n is thought to arise from low density and low crystallinity of each film owing to their low processing temperature. Further, the measured extinction coefficients of SiO₂, Nb₂O₅, SnO₂ and Ag are 0.02, 0.0, 0.0 and 2.98, respectively.

Prior to experiments, the optical programs named Essential Macleod Program (EMP) was carried out to simulate the optical characteristics. Fig. 3(a) exhibited the optical simulation results of the transmittance on the SnO₂ (40 – 55 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (20 - 35 nm) multi layer as a function of SnO₂ thickness.

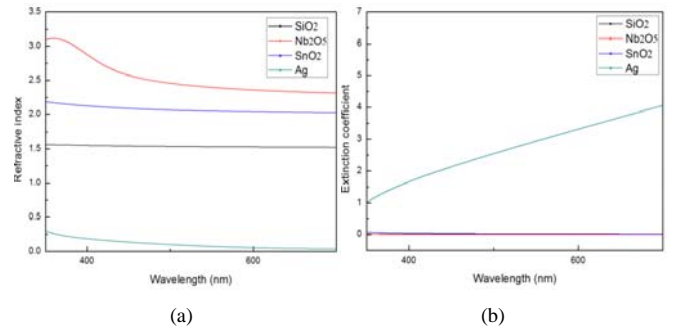


FIGURE II. (A) THE MEASURED REFRACTIVE INDEX n AND (B) EXTINCTION COEFFICIENT k FOR SiO₂, Nb₂O₅, SnO₂ AND AG THIN FILM IN VISIBLE RANGE

Fig. 3(b) shows the experimentally measured transmittance of the SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ multilayer films as a function of the SnO₂ thickness. The transmittance of the SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ film with the bottom thickness of SiO₂ 30 nm was 85.4 % at 550 nm wavelength. As compared with simulation data, transparency obtained from experimental results was relatively lower than that obtained through simulation for wavelength values above 550 nm. This might be attributed to light scattering on the each layer of film surface and interface instability due to diffusion. Moreover, the substrate temperature was also considered to cause differences between the experimental and EMP simulation results.

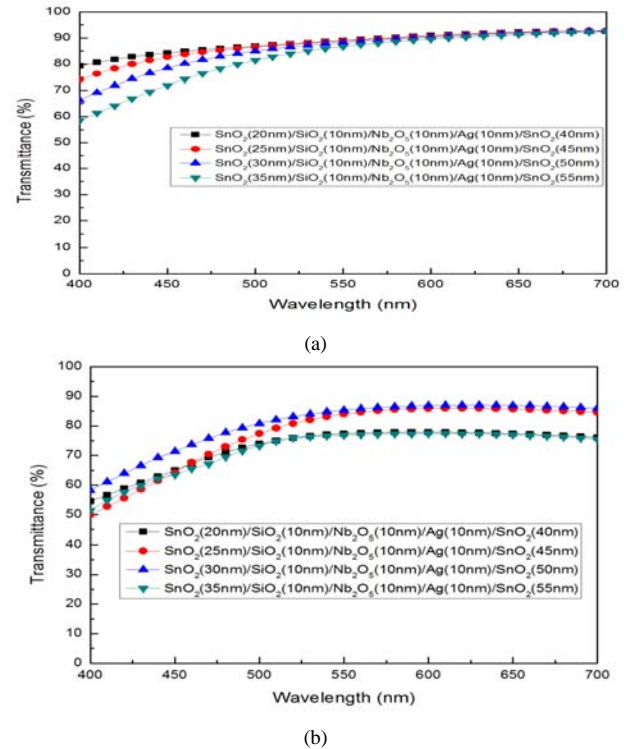


FIGURE III. (A) SIMULATED AND (B) EXPERIMENTALLY MEASURED OPTICAL TRANSMITTANCE SPECTRA ON THE SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ MULTI LAYER FILM AS A FUNCTION OF THICKNESS OF THE TOP AND BOTTOM SnO₂ LAYER

Based on the results taken from Fig. 3, the film thickness of the top SnO₂ layer varied from 40 to 55 nm, whereas the SiO₂,

Nb₂O₅ and Ag layers were fixed at 10 nm and the bottom SnO₂ layer was kept at 30 nm. Fig. 4 demonstrates the transmittance spectra from simulation and experiment taken on the SnO₂ (40 – 55 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm), respectively. As can be seen in simulation spectra of Fig. 4(a), the transmittance gradually decreased with increasing the top SnO₂ thickness from 40 to 55 nm. The transmittance of the multilayer films had in the range of 86.3 – 89.7%, high transparency in the visible range of 550 nm. Fig. 4(b) shows the experimentally measured results of the transmittance on the same film. It is noted that the transmittance window narrows and gradually becomes lowered relatively, as compared with simulation results. However, the experimental transmittance of SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ thin films was in the range of 84.3 – 85.8 at 550 nm, slightly different regardless of the top SnO₂ thickness. It was shown that the SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) film yielded the highest optical transmittance of 85.8 % at 550 nm.

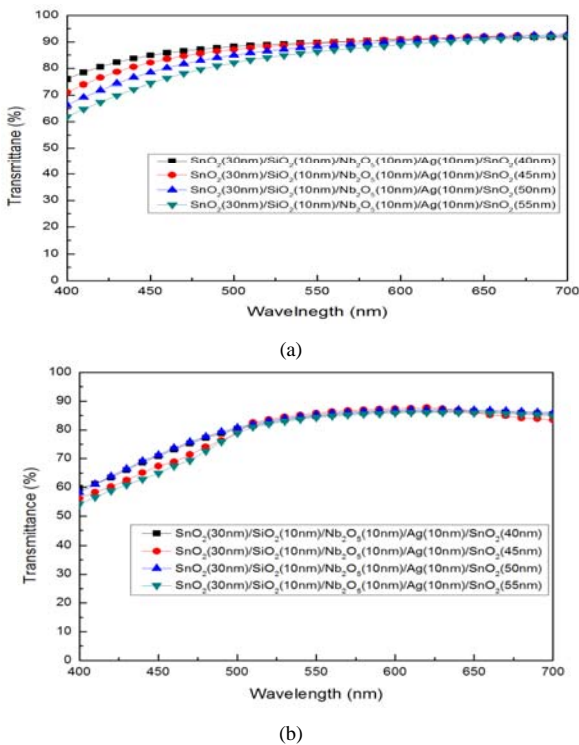


FIGURE IV. (A) SIMULATED AND (B) EXPERIMENTALLY MEASURED OPTICAL TRANSMITTANCE SPECTRA ON THE SnO₂ (40–55 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) MULTI LAYER FILM

B. Phase Identification and Surface Morphology Analysis

Fig. 5 presents the XRD patterns obtained from the SnO₂ (40 – 55 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) multi layer film with various SnO₂ layer thicknesses. As can be seen, all of the as-deposited multi layer films appear to be amorphous. Regardless of the SnO₂ thickness, there is a broad at 26° for all samples that is caused by the glass substrate. This can be attributed to the fact that thin film with about 100 nm thick has a nanocrystalline phase formed during DC/RF sputtering process. Another feature seen in the Fig. 5 is that Ag (111) peaks could be observed around 38.2 after deposition Ag on SnO₂/SiO₂/Nb₂O₅ multi layer film.

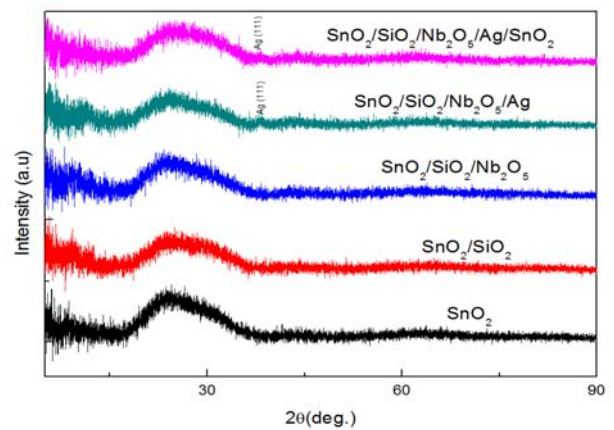


FIGURE V. XRD PATTERNS OBTAINED FROM THE SnO₂ (40 – 55 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) MULTI LAYER FILM WITH VARIOUS SnO₂ LAYER THICKNESSES

The surface morphologies of the Ag films as a function of Ag layer thicknesses are shown in Fig. 6. The Ag particles begin to connect to each other and start forming a continuous path at the thickness of around 10 nm. It was known that the total resistivity of the multi layer film is mainly determined by Ag layer, since the resistivity of Ag is much lower than that of SnO₂.

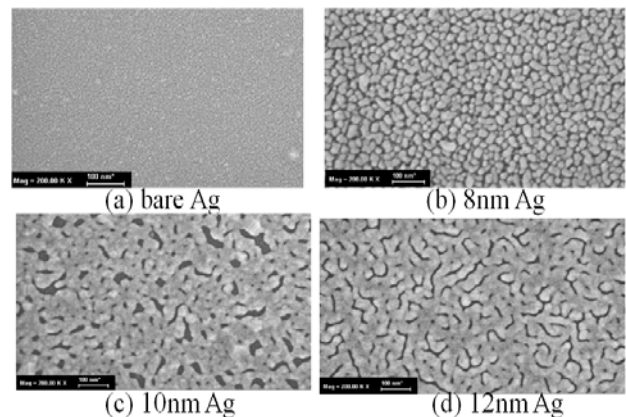


FIGURE VI. SURFACE MORPHOLOGIES OF THE AG FILMS AS A FUNCTION OF AG LAYER THICKNESS

Fig. 7 shows the surface roughness of the deposited films was investigated using AFM. Measurement has been taken on 5 different multi layer films continuously deposited, (a) SnO₂ (45 nm), (b) SnO₂ (45 nm)/Ag (10 nm), (c) SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm), (d) SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm) and (e) SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm). It can be seen that the surface roughness of the SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) film in Fig. 7 was related to that of the underlying index matching layer. As seen in Fig. 7, the roughness increased after SnO₂ layer was deposited. However, surface roughness maintained are relatively small in the range of 4.0 nm, after deposition of Nb₂O₅ and SiO₂ index matching layers.

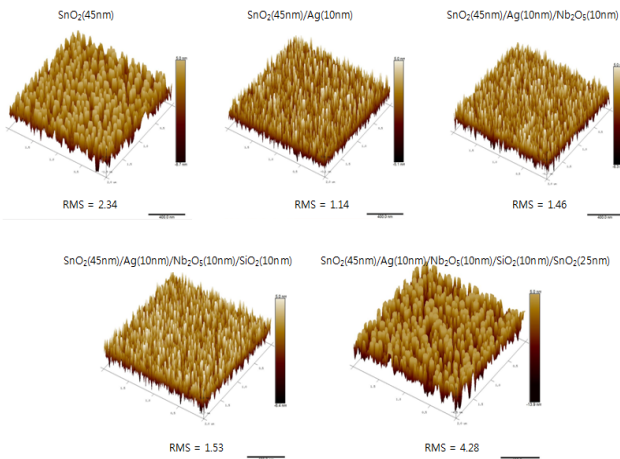


FIGURE VII. AFM IMAGES OF SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) Multi Layer Film

Fig. 8 show the AES depth profiles of the SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) multi layer. All of the peaks in the spectra can be accounted for by the components of the film. The depth profiling spectra shown in Fig. 8 seems to show that interface between each layers was not well defined. Sn, Si and Nb fractions are diffuse within the explored depth and especially oxygen element was detected clearly on the silver matrix. One possible explanation is that there was a possible interfacial reaction between layers and thereby, element of the each layer has diffused into the film during processing.

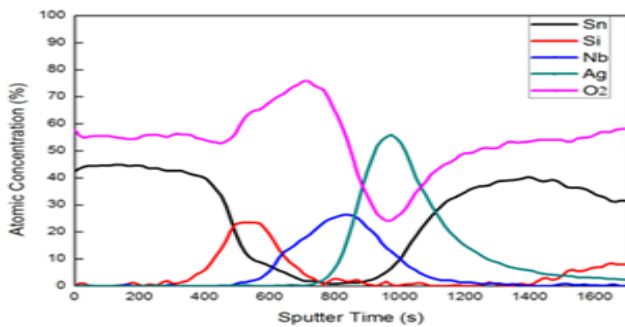


FIGURE VIII. AES DEPTH PROFILES OF THE SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) MULTI LAYER FILM

IV. CONCLUSION

In summary, the characteristics of the RF/DC sputtering grown hybrid structure of SnO₂/Ag/Nb₂O₅/SiO₂/SnO₂ with varying the SnO₂ thicknesses were systematically investigated. In order to estimate the optical characteristics and compare them with experimental results, EMP was adopted and the refractive index n and extinction coefficient k were measured. The transmittance and transmittance were dependent on the SnO₂ thickness. The measured transmittance suggested that the multilayered thin film of SnO₂ (45 nm)/Ag (10 nm)/Nb₂O₅ (10 nm)/SiO₂ (10 nm)/SnO₂ (30 nm) exhibited high transmittance of 85.8 % at 550. The XRD patterns obtained from the SnO₂ /Ag/Nb₂O₅ /SiO₂ /SnO₂ multi layer film appear to be amorphous. The depth profiling spectra show that interface

between each layer was not well defined, indicating there was a possible interfacial reaction between layers.

V. ACKNOWLEDGEMENT

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