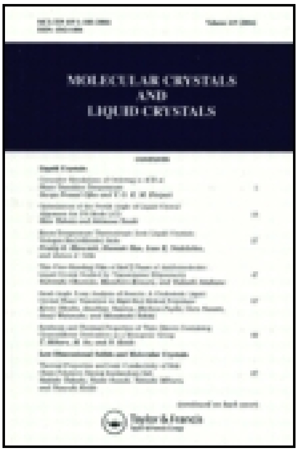

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Characteristics of AZO/Cu/AZO Multilayer Thin Films Prepared on Polyethersulfone Substrate at Room Temperature

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Highly conducting Al doped ZnO(AZO)/Cu/AZO triple multilayer thin films were deposited on polyethersulfonesusbtrate at room temperature. We investigated the structural, electrical, optical, and mechanical properties of triple multilayers as a function thickness of Cu layers. The triple multilayer with flexible substrate had advantages such as low sheet resistance and stable mechanical properties as compared with single oxide layer. From the results, sheet resistance value of AZO(50 nm)/Cu(9 nm)/AZO(50 nm) multilayer was 12 Ω/□, and average optical transmittance(380–770 nm) value of multilayer was 80%. Moreover the triple multilayer showed mechanical flexural strength properties than single-layered AZO thin films during bending test due to the existence of ductile Cu metal layer.

Keywords AZO/Cu/AZO; FTS; Triple multilayer

Introduction

Transparent conductive-oxide (TCO) thin films have been deposited on a polymer substrate that has light weight, small volume, flexibility, and portability. The TCOs with polymer substrate are widely used in flexible displays, electro-optical devices, and thin-film solar cells [1–3]. Al-doped ZnO is a promising TCO material because it is nontoxic, inexpensive, and stable in hydrogen plasma, as compared with indium tin oxide (ITO) [4]. However, the resistivity of Al-doped ZnO is not low enough, in some cases, for improved applications. In order to enhance the conductivity of transparent conducting films, it is necessary to experiment withnew materials [5]. It is well known that the conductivity of metal films is very high, but their optical transmittance in visible range is relatively low [6]. Fan et al. [7] reported that the dielectric/metal/dielectric-multilayer film could suppress the reflection from the metal in the visible region and that it achieved a selective transparent effect. Recently, triple multilayer structures, such as TCO/metal/TCO, have shown their usefulness in transparent conducting materials. Moreover, the triple multilayers structures exhibited stable mechanical properties compared to single-layered TCO samples during the bending test because of the existence of a ductile metal layer in triple layered film.

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The metals for practical usesin thin films are Al, Cu, and Ag, however, when compared with Cu, Ag is more expensive, and Al is more sensitive to oxygen;therefore, we selected a Cu metal layer. In this work, we will investigate an AZO/Cu/AZO triple multilayer as a function of the thickness of an intermediate Cu metal layer grown on a polyethersulfone substrate using the facing targets sputtering (FTS) method.

Experimental

Figure 1 shows aschematic of the FTS method used in the triple multilayer thin film deposition process. The FTS method was designed to array the two targets so as to face each other, and to the form high-density plasma between the targets. The FTS system was able to restrain the bombardment of the substrate by high-energy particles because the substrate's position was located outside of the plasma. Consequently, the FTS system suppressed the substrate damage caused by high-energy particles, such as electrons and partial ions[4,8]. In this work, Cu targets were installed on top and AZO targets were installed on the bottom. The chamber was evacuated to 1.6×10^{-4} Pa before the film deposition began, using a rotary pump and a turbo molecular pump. Before each run, the target was pre-sputtered in a pure argon atmosphere for 10 min in order to remove the natural surface oxide layer of the Cu, and the AZO targets. The polyethersulfone substrate was ultrasonically cleaned using isopropyl alcohol (IPA) and DI-water for 30 min and blown dry with N_2 gas. More details about the sputtering conditions are given in Table 1. Thicknesses of triple multilayer thin films were measured by using a surface profiler (alpha-step). Electrical properties of the thin films were measured by using a four-point probe. Optical transmittance and structural properties were measured by using a UV/VIS spectrometer, an X-ray diffractometer, and

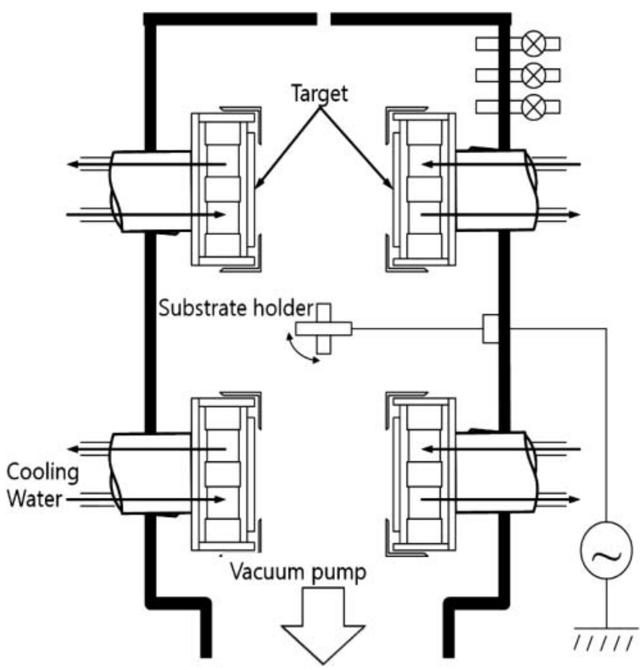


Figure 1. Schematic of FTS method used in the triple multilayer thin film deposition process.

Table 1. Sputtering conditions

Deposition Parameter	Sputtering Conditions	
Targets	AZO ($ZnO : Al_2O_3 = 98: 2$ wt.%, 4N, 4inch)	Cu(4N, 4 inch)
	AZO ($ZnO : Al_2O_3 = 98: 2$ wt.%, 4N, 4inch)	Cu(4N, 4 inch)
Substrate	Polyethersulfon (PES)	
Base pressure	2.6×10^{-4} Pa	2.6×10^{-4} Pa
Working gas pressure	0.13 Pa	0.13 Pa
Film thickness	Top: 50 nm, Bottom: 50 nm	3–18 nm
Substrate temperature	Room temperature	Room temperature
Input power	85 W	15 W
Working gas	Ar 12 sccm	Ar 12 sccm
Deposition rate	8 nm/min	2.7 nm/min

field emission scanning electron microscopy (FE-SEM). Depth profiling of the materials in the triple multilayer were measured by using Secondary Ion Mass Spectrometry (SIMS). Finally, the mechanical flexural strength of the single-layered AZO and the triple multilayer thin films were measured by using a bending tester.

Results and Discussion

The electrical and optical properties of triple multilayerthin filmsshould be excellent. Therefore, to confirm this, we measured their sheet resistance and optical transmittance in the visible range. Figure 2 shows the sheet resistance and average optical transmittance in

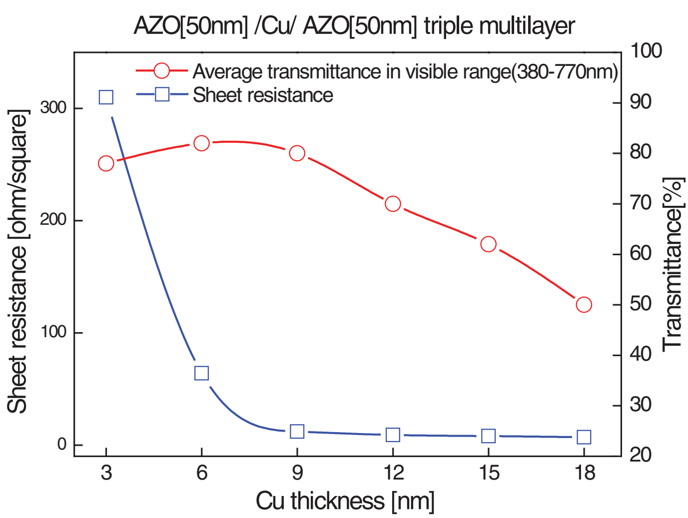


Figure 2. Sheet resistance and average optical transmittance in visible range of triple multilayer as a function of intermediate Cu metal layer.

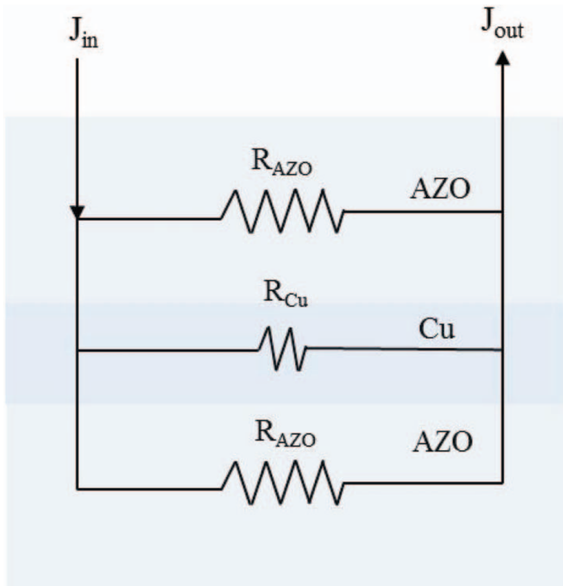


Figure 3. Schematic of parallel circuit of three resistors (AZO/Cu/AZO).

the visible range of triple multilayerthin films as a function of the intermediate Cu metal layer. As the Cu metal layer's thickness increased,the sheet resistance decreased.The sheet resistance of the multilayer showed 310, 64, 12, 9, 8, and 7 Ω/□ with 3, 6, 9, 12, 15, and 18 nm thicknesses, respectively.The average optical transmittance in the visible range of the single layered AZO thin films exhibited was 93%. The average optical transmittance of the multilayer thin films in the visible range showed 78, 82, 80, 70, 62, and 50 Ω/□with 3, 6, 9, 12, 15, and 18 nm thicknesses, respectively. The sheet resistance and average transmittance depended on the thickness of the intermediate Cu metal layer because the Cu layer changed from discontinuous film to continuous film [9]. Figure 3 shows a schematic of a parallel circuit of three resistors (AZO/Cu/AZO). The triple multilayer thin films can be considered as a parallel circuit of three resistors [10]; so, the relationship among the sheet resistance of the triple multilayer thin film (R_S), AZO layer (R_{AZO}), and intermediate Cu metal layer (R_{Cu}) is followed by:

$$\frac{1}{R_s} = \frac{1}{R_{AZO}} + \frac{1}{R_{Cu}} + \frac{1}{R_{AZO}} \tag{1}$$

But, the conductivity of the triple multilayer thin films is mainly supplied by the metal layer resistance [11].

Figure 4 showsthe optical transmittance in the visible range of the triple multilayer thin films. As the intermediate Cu metal layer increased from 3 nm to 6 nm, the average transmittance increased from 78% to 82%, and when the Cu thickness was increased from 6 nm to 12 nm, the transmittance decreased. As discussed above, when the Cu layer is very thin, it is discontinuous [12], which leads to the diffuse reflection of incident light, resulting in the low transmittance. When an appropriate thickness (6 nm and 9 nm in our case) is obtained, the Cu layer was continuous. [13]. Figure 5 shows thetransmittance in the visible range of the single-layered Cu metal film as a function of Cuthickness. While the thickness

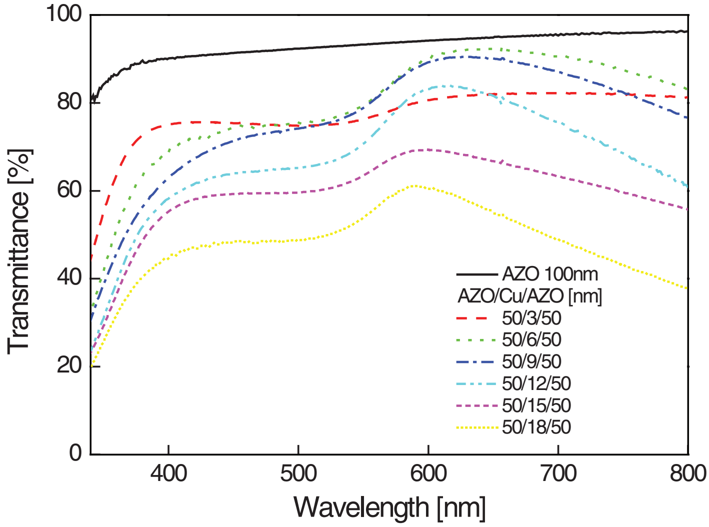


Figure 4. Optical transmittance in visible range of triple multilayer thin films.

of the single-layered Cu metal thin film increased, the average optical transmittance of the single-layered Cu metal thin film decreased. Generally, the optical transmittance of the triple multilayer thin film was closely related to the intermediate Cu metal layer and the top transparent conductive oxide. The top TCO thin-film layer prevents the intermediate Cu metal layer from turning into an oxidized Cu layer and plays a role in the antireflection layer blocking the reflected light [14]. Therefore, the optical transmittance was decreased by light absorption, scattering, and reflection [15].

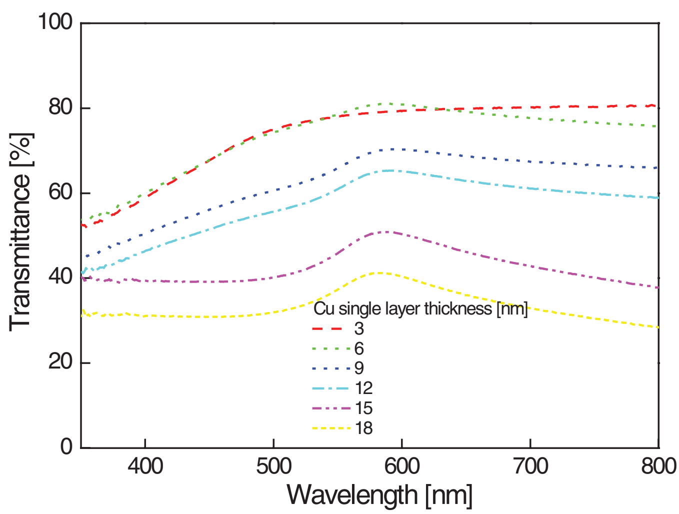


Figure 5. Optical transmittance in visible range of the single-layered Cu metal film as a function of Cuthickness.

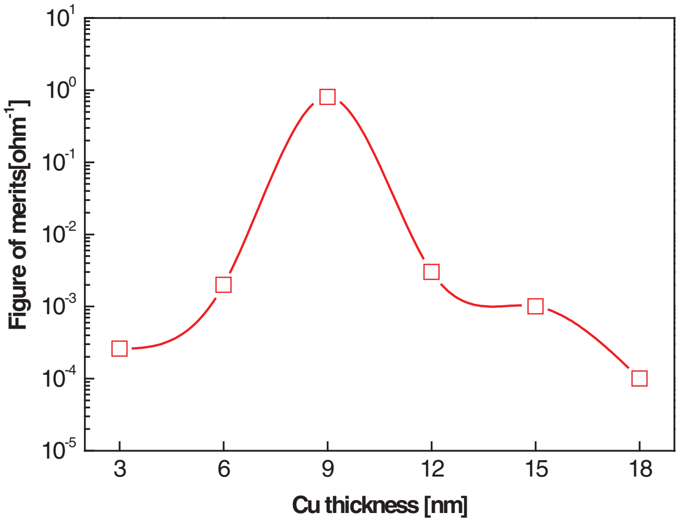


Figure 6. The figure of merits suggested by Haacke [16] as a function of the Cu metal thickness.

Figure 6 shows the figure of merits (=FOM) suggested by Haacke [16] as a function of the Cu metal thickness. In various applications of transparent conductive films, the optical and electrical properties of the films are very important. Haacke [16] proposed a = FMO followed by:

$$\phi_{TC} = \frac{T^{10}}{R_{Sh}} \tag{2}$$

where T is the average optical transmittance and R_{sh} is the electrical sheet resistance of the TCOs. This equation can be used to compare the performance of transparent conductive films. The maximum value ($= 0.8 \times 10^{-2}\ \Omega^{-1}$) was found when the Cu metal thickness was 9 nm (T: 0.8 and R_{sh}: 12 Ω/□). The higher value of the triple multilayer compared to the single-layered AZO indicates that AZO/Cu/AZO is a high performance electrode. Figure 7 shows the [ED: Spell out the abbreviation.]XRD peak of the triple multilayer thin films as a function of Cumetal thickness.It is observed that all the films are polycrystalline with the (002) preferred orientation of ZnO hexagonal wurtzite. For the film with a Cu thickness from 3 nm, the Cu peak was not observed. The surface of the Cu layer was not continuous and the quality was poor. For each of the other films, the Cu(111) peak was detected, and the intensity of peak was enhanced with the increase of Cu thickness. Figure 8 shows a surface SEM image of the Culayer located on the bottom AZO layer as a function of the Cu thickness. It shows that the morphology of the Cu metal layer changed with an increasing Cu thickness. When the Cu layer is very thin (3 nm), it is discontinuous. When the Cu layer increased to 6 nm, the surface morphology transforms to a compact and smooth structure. The smooth structure can reduce the scattering of incident light, which makes some contribution to the increase of the transmittance [17]. The mechanical flexural strength of TCO films on a flexible substrate should be excellent; therefore, to confirm this, we measured mechanical properties of the triple multilayer on a flexible substrate. Figure 9 shows the mechanical flexural strength of the single-layered AZO and triple multilayer. A total of 300 cycles were iterated, and the resistance of the single-layered AZO thin film

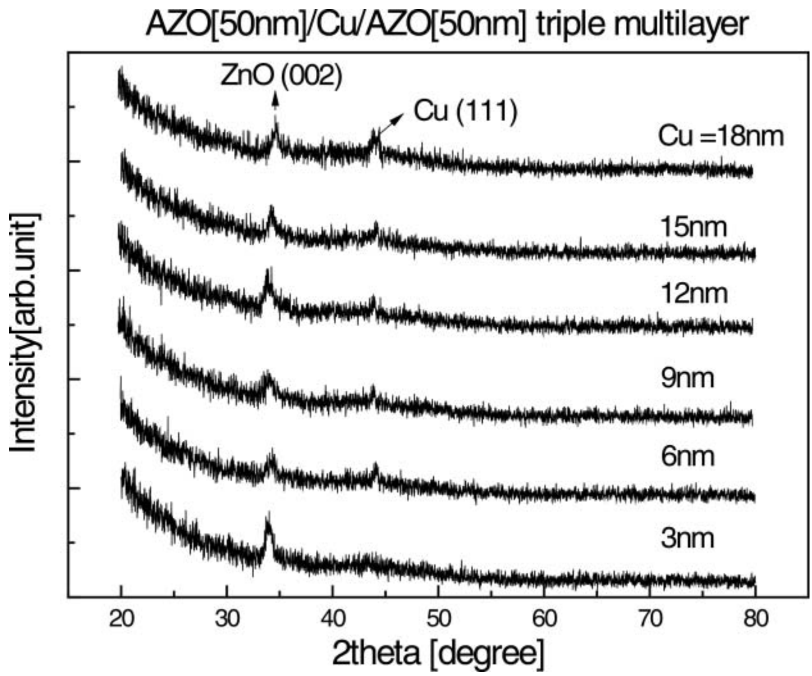


Figure 7. XRD peak of the triple multilayer thin films as a function of Cumetal thickness.

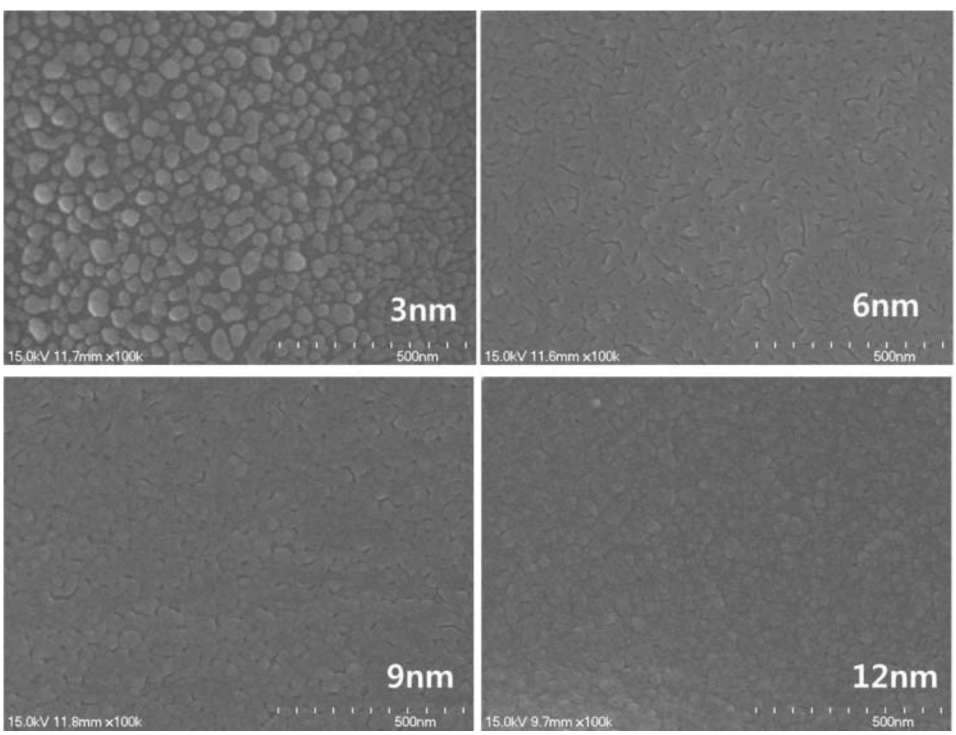
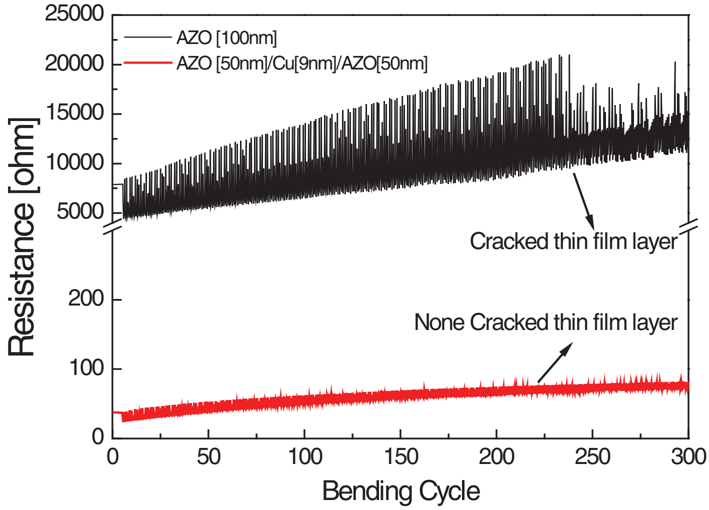
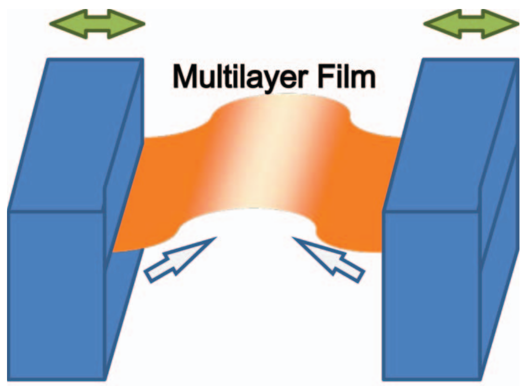


Figure 8. Surface SEM image of the Culayer located on the bottomAZO layer as a function of the Cu thickness.

(a)



(b)



(c)

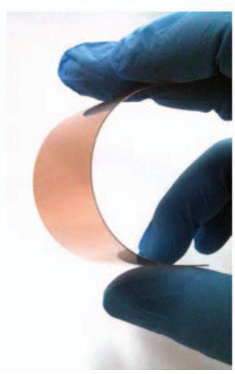


Figure 9. (a) Mechanical flexural strength of single-layered AZO and triple multilayer. (b) Schematic of bending tester. (c) Photograph of AZO/Cu/AZO triple multilayer.

rapidly increased above 240 cycles, and the resistance of the triple multilayer was fixed. Moreover, the triple multilayers showed more stable mechanical properties than the single-layered AZO sample did during the bending test because of the existence of the ductile Cu metal layer.

Conclusions

In this work, electrical, structural, optical and mechanical properties of AZO/Cu/AZO triple multilayer structure on polyethersulfone substrate were investigated as a function of Cu layer thickness. 9 nm of Ag metal layer thickness, sheet resistance and average optical transmittance in visible range of triple multilayer thin film 12 Ω/□ and 80%, and figure of merits exhibited $0.8 \times 10^{-2}\ \Omega^{-1}$. Moreover the triple multilayers showed stable mechanical properties than single-layered AZO sample during the bending test due to the existence of ductile Cu metal layer. The AZO/Cu/AZO triple multilayer thin films could apply flexible transparent electrodes.

Acknowledgment

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References

[1] Yang, T. L., Zhang, D. H., Ma, J., Ma, H. L., & Chen, Y. (1998). *Thin Solid Films*, *326*, 60–62.
[2] Moon, C. S., Chung, Y. M., Jung, W. S., & Han, J. G. (2007). *Surf. Coat. Technol.*, *201*, 5035–5038.
[3] Fortunato, E., Nunes, P., Costa, D., Brida, D., Ferreira, I., & Martins, R. (2002). *Vacuum*, *64*, 233–236.
[4] Jung, Y. S., Park, S. J., Yoon, H. H., Choi, H. W., & Kim, K. H. (2009). *J. Korean Phys. Soc.*, *55*, 1945–1949.
[5] Igasaki, A. Y., & Kanma, H. (2001). *Appl. Sur. Sci. 169–170*, 508–511.
[6] Choi, K. H., Kim, J. Y., Lee, Y. S., & Kim, H. J. (1999). *Thin Solid Films*, *341*, 152–155.
[7] John C.C. Fan, Thomas B. Reed, & John B. Goodenough (1974). SAE Preprints, in: Proceedings of the Ninth Intersociety Energy Conversion Engineering Conference, vol. 26–30, pp. 341.
[8] Matsushita, N., Noma, K., Nakagawa, S., & Naoe, M. (1992). *ICF*, *6*, 428–431.
[9] Park, H. K., Kang, J. W., Na, S. I., Kim, D. Y., & Kim, H. K. (2009). *Sol. Energy Mater. Sol. Cells*, *93*, 1994–2002.
[10] Klöppel, A., Kriegseis, W., Meyer, B. K., Scharmann, A., Daube, C., Stollenwerk, J., & Trube, J. (2000). *Thin Solid Films*,*365*,139–146.
[11] Bender, M., Seelig, W., Daube, C., Frankenberge, H., Ocker, B., & Stollenwerk, J. (1998). *ThinSolid Films*, *326*, 67–71.
[12] Liu, X., Cai, X., Qiao, J., Mao, J., & Jiang, N. (2003). *Thin Solid Films*, *441*, 200–206.
[13] Sahu, D. R., Lin, S. Y., & Huang, J. L. (2008). *Thin Solid Films*, *516*, 4728–4732.
[14] Fahland, M., Karlsson, P., & Charton, C. (2001). *Thin Solid Films*, *392*, 334–337.
[15] Yamamoto, M., & Namioka, T. (1992). *Appl. Opt.*, *31*, 1622–1630.
[16] Haacke, G. (1976). *J. Appl. Phys.*, *47*, 4086–4089.
[17] Sahu, D. R., & Huang, J. L. (2006). *Mater SciEng, B*, *130*, 295–299.