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Oxide/metal/oxide electrodes for solar cell applications

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ABSTRACT

In this paper the influence of the heat treatment on the structural and optical properties of oxide single-layer and multilayer transparent thin film structures of ITO/Au/ITO, AZO/Au/AZO, $TiO_2/Au/TiO_2$ and $Bi_2O_3/Au/Bi_2O_3$ type is studied. The single-layer oxides and respective multilayer structures have been deposited on glass substrates by successive DC magnetron sputtering using metallic targets In:Sn, Zn:Al, Ti and Bi, in reactive (for the oxide films) or inert (for the metallic Au interlayer films) atmosphere. Good quality transparent conducting thin film structures have been obtained, with resistivity $\sim 10^{-4}\ \Omega$ cm and transmittance ~75%. The wetting surface properties in function of time exposure at UV radiation (254 nm) have been also studied before and after heat treatment performed at 450 °C.

1. Introduction

In recent years a significant efficiency improvement of the organic (OPV) and hybrid photovoltaic devices exceeding 10% (Adebanjo et al., 2013) and 21% (Park, 2013; Green et al., 2014), respectively has been obtained at laboratory scale. Large scale modules with efficiencies comparable to those of small scale devices and manufactured with lower cost materials are needed to develop industrial and commercial applications. Typically large scale OPV and hybrid modules are 20–30% less efficient than the corresponding single cells. One of the main reasons of the efficiency drop is related to the high sheet resistivity of the Indium Tin Oxide (ITO) transparent electrode at large scale (Pandey and Nunzi, 2011). The ITO is at the moment the most used material as transparent electrode (Girtan, 2005; Girtan and Mallet, 2014; Granqvist, 2007, 2003). However, the limited resources of Indium on earth and the expensive cost of this material require reducing the amount of material employed for thin film fabrication or even the replacement of ITO by other materials displaying equivalent properties (Girtan et al., 2006, 2009, 2013, 2010; Rusu et al., 2008; Ghomrani et al., 2011; Socol et al., 2016; Koralli et al., 2016; Manole et al., 2013; Saidi et al., 2016). The thickness of ITO thin films used as transparent electrodes for optoelectronic devices ranges in general, between 150 and 700 nm. The reduction of single-layer ITO film thickness below 150 nm is not possible, because of the increase in the electrical resistivity with the decreasing thickness (the classical size effect of semiconductors).

In our previous studies (Girtan, 2012), in 2012, we indicated that ITO/Metal/ITO (IMI electrodes) and ZnO/Metal/ZnO thin-film structures prepared on glass and plastic substrates, by reactive sputtering from metallic targets, are very promising electrodes for the new generation solar cells and transparent plastic electronics. The figure of merit is as high as the one of ITO single layers but the amount of ITO is reduced by 4. Moreover, the stability of the mechanical and electric properties is remarkable in comparison with the usual single-layer oxide films.

Following that, in 2014, the group of Brabec realized for the first time an OPV module using ITO/Metal/ITO electrodes (Kubis et al., 2014), as described in Girtan (2012), and in 2015 BELECTRIC OPV presented, at World Exhibition of Milan, large scale OPV modules on plastic substrates using IMI electrodes (Berny et al., 2016).

Encouraged by the previous results and the subsequently large scale fast applications, we continued our studies on the ITO/Au/ITO and AZO/Au/AZO and also on other types of Oxide/Metal/Oxide multilayer structures, such as $TiO_2/Au/TiO_2$ and $Bi_2O_3/Au/Bi_2O_3$.

Titanium dioxide (TiO_2) is an intensively studied oxide (Mardare et al., 2010; Iftimie et al., 2009; Vaiciulis et al., 2012), for its relatively low cost, simple manufacture and for the high-performance of dye-sensitized (DSSC) and perovskite solar cells (Green et al., 2014; Hagfeldt et al., 2012; Nazeeruddin and Grätzel, 2003; Millington, 2009; Burschka and et al., 2013; Lee

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Table 1
Sample deposition conditions.

Layer	Atmosphere conditions	Target - substrate distance (cm)	Deposition current (mA)	Pressure (10^{-2} mbar)	Deposition time	Target composition (wt%)
ITO	Reactive atm.	7	30	2	4 min	In 90%, Sn 10%
Au	Argon atm.	7	30	1	14 s	Au 100%
ITO	Reactive atm.	7	30	2	4 min	In 90%, Sn 10%
AZO	Reactive atm.	7	100	2	4 min	Zn 98%, Al 2%
Au	Argon atm.	7	30	1	14 s	Au 100%
AZO	Reactive atm.	7	100	2	4 min	Zn 98%, Al 2%
TiO_2	Reactive atm.	7	30	2	4 min	Ti 100%
Au	Argon atm.	7	30	1	14 s	Au 100%
TiO_2	Reactive atm.	7	30	2	4 min	Ti 100%
Bi_2O_3	Reactive atm.	7	30	2	4 min	Bi 100%
Au	Argon atm.	7	30	1	14 s	Au 100%
Bi_2O_3	Reactive atm.	7	30	2	4 min	Bi 100%

Table 2
Thin-film sample thickness.

Sample	*d* (nm)
ITO	45
ITO/Au/ITO	45/7/45
AZO	40
AZO/Au/AZO	40/7/40
TiO_2	30
TiO_2/Au/TiO_2	30/7/30
Bi_2O_3	120
Bi_2O_3/Au/Bi_2O_3	120/7/120

et al., 2012) based on it. Here the titanium oxide film doesn't act as an electrode, but as an active layer in a process similar to the photosynthesis (Lewis, 2004; Pandit and Frese, 2012). The classical architecture is FTO (Fluorine doped Tin Oxide)/TiO_2/Perovskite/HTL (hole-transport layer)/Au. The TiO_2 layer on FTO glass substrates is usually deposited by sintering a titania paste at high temperature (500 °C). The replacement of FTO by a TiO_2/Metal/TiO_2 (TMT electrode) could have at least two advantages: 1) the deposition of electrodes on large area flexible substrates at low tempera-

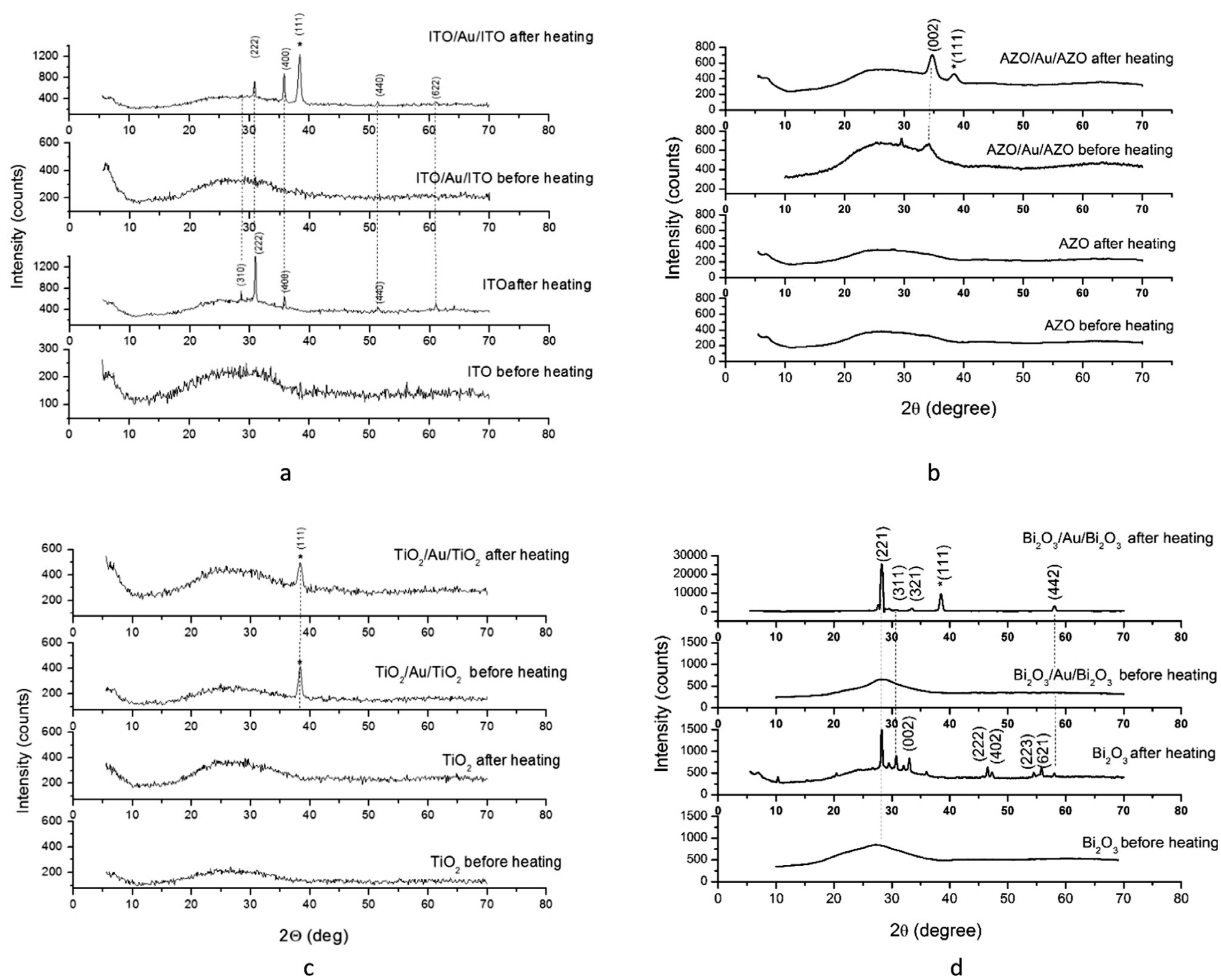

Fig. 1. XRD-ray diffraction patterns of oxide single layers and oxide/metal/oxide multilayer structures as deposited and after the heat treatment at 450 °C.