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Growth and fabrication of dye sensitized solar cells on multilayer transparent conductive films

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Dye sensitized solar cells (DSSC) are of great interest for the conversion of solar energy into electrical energy and are future alternatives to silicon solar cells. We have synthesized doped ZnO and Ag multilayer which is used as transparent electrode. Small quantity of ZnO incorporated TiO_2 matrix is used for the fabrication of DSSC. Chemical vapour deposition method is used to prepare ZnO covered TiO_2 film. The DSSC on the Al doped ZnO and Ag multilayer covered with ZnO/TiO_2 film yielded an overall cell efficiency of 5.45 %. Therefore, DSSC using low cost transparent conducting oxide film and mercurochrome dye is a nice step to develop DSSC.

1. Introduction

A lot of efforts have been taken to explore the possibilities of fabricating highly efficienct dye sensitized solar cells (DSSC) with low price [1-5] which is an alternative to silicon solar cells. The transparent electrode with maximum collection of electron and fast electron movement in the transport medium is a requirement to improve the efficiency of DSSC [6]. This problem can be overcome with the use of transparent electrode having high conductivity and transmittance. Multilayer transparent conductive (TCO) electrode consist of dielectric /metal/ dielectric can be fabricated [7,8] which can maintain high conductivity, low reflectance and good transparency. In DSSC, TiO_2 particles are deposited on the conducting electrode and heated about 450-500°C for efficient electronic contact [9]. Generally, the conductivity of conducting glass decreases significantly after heating at this temperature [10]. Further, the DSSC with Indium tin oxide (ITO) substrates not show significant enhancement of efficiency due to increase of resistivity during fabrication. In order to achieve high performance DSSC along with stability on the performance of DSSC, there is a need for development of highly conductive, high transmittance and heat stability conducting substrate.

This work reports the efficient DSSC which is fabricated using highly conducting multilayer transparent electrodes and low temperature sintered electrodes of mesoporous TiO_2. Multilayer Al doped ZnO (AZO) and Ag layer are fabricated as AZO/Ag/AZO transparent conductive films and their application to DSSC was examined. Further, a ZnO covered TiO_2 electrode was also prepared using chemical vapor deposition (CVD) and its photovoltaic properties are studied. In addition, much cheaper mercurochrome ($C_{20}H_8Br_2HgNa_2O$) dyes is used as photosensitizer [11].

2. Experimental Method

Nanocrystaline TiO_2 prepared using the previous procedure [12] was used for the fabrication of DSSC on prepared AZO/Ag/AZO (named as AAA) TCO substrate. Detail procedures for the preparation of AAA multilayers are described elsewhere [13]. The thickness of multilayer AAA transparent conductive glass is 25 /11/25 nm based on our reported work [13]. Further, to get the best possible TCO, more AAA with different thickness of upper surface layer are fabricated varied from 0 to 100 nm. Transmittance spectra were recorded using UV-VIS to know the optical properties of the film. The titanate precursor are prepared using 3 gm of a TiO_2 powder (P25, Degussa) mixed with 100 ml of 10 N NaOH in an autoclave at 130^0C for 20 h. Then, it is washed with 0.1 N HNO_3 to get pH value of 1.5. The TiO_2 suspension for DSSC was obtained by autoclaving the titanate suspension at 240^0C for 12 h. The dispersion was then deposited uniformly onto the multilayer TCO to form a TiO_2 film. The obtained film was calcined at 450^0C for 30 min to form a mesoporous electrode. For ZnO covered TiO_2 electrode, ZnO film was deposited using CVD process. The Zn power is kept in a ceramic boat and placed inside the quartz tube of the furnace. The powders are heated slowly by increasing the temperature of the furnace to 500°C in argon atmosphere. The ZnO deposited on TiO_2 film was impregnated with dye at room temperature. Then DSSC was fabricated.

The DSSC configuration was as follows: glass/multilayer TCO/ TiO_2 / Mercurochrome dye/ liquid electrolyte/ Pt / ITO / glass. The dye absorption was performed for 3 minutes on the TiO_2 electrodes once it is heated at 450^0C for 30 min and cooled. The dye absorbed TiO_2 electrode were used to make a sandwich type cell. Pt-sputtered ITO glass is used as the counter electrode. Photocurrent-voltage measurement were performed using AM 1.5

solar simulator (100 mW cm^{-2}). The measurement on active area of the DSSC was about. 0.5 cm^2.

3. Results and Discussion

The transmission spectra of AAA multilayer along with ITO and FTO are presented in Figure 1. It is observed that the transmittance of AAA multilayer with upper surface layer of 25 nm is comparable with that of ITO and FTO at the wavelength range of 500 to 550 nm with good conductivity (7 Ω/Sq.) The transmittance of AAA is also above 80 % which is suitable for DSSC as an alternative to FTO and ITO. Further the transmittance spectra of other multilayer with different thicknesses are presented here for comparison.

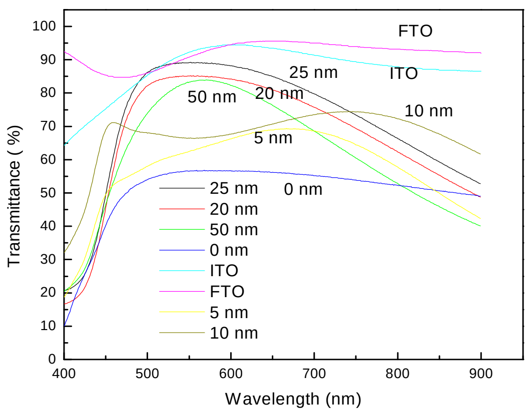

Figure 1. The transmission spectra of multilayer with AZO surface layer of thickness from 0 to 50 nm along with ITO and FTO.

Figure 2 presents the variation of transmittance and reflectance of multilayer TCO with thickness of the surface AZO layer in the wavelength range of 400-700 nm. The average maximum value of transmittance and minimum value of reflectance is observed for coating of 20-25 nm AZO. The overall changes in the value of transmittance and reflectance occur after 50 nm thickness of AZO surface layer. This is due to the phenomenon of interferences in the surface AZO layer. The maximum transmittance of about 0.86 is achieved in the visible range for AAA with thickness of 25 nm/11 nm/25 nm

Using developed AAA, the performance of DSSC is measured after deposition of mesoporous TiO_2. Figure 3 indicates the light to energy conversion efficiencies (n) of the DSSC with the thickness of the mesoporous TiO_2 layer. The results show that the n value was 2.26 % for single coating cycle of TiO_2. This n value increases with deposition of more layer of TiO_2. The maximum value of 5.07 % was achieved at four coating cycles.

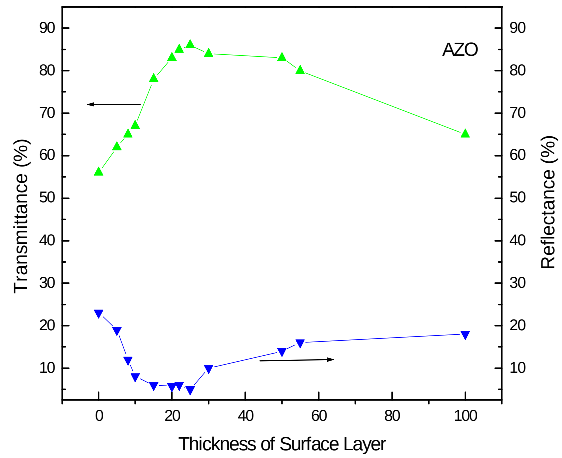

Figure 2 The reflectance and transmittance spectra of multilayer AAA with different thickness of the AZO surface layer

It is known that there must be an optimum thickness of TiO_2 on the coating to achieve the maximum efficiency [14-16].

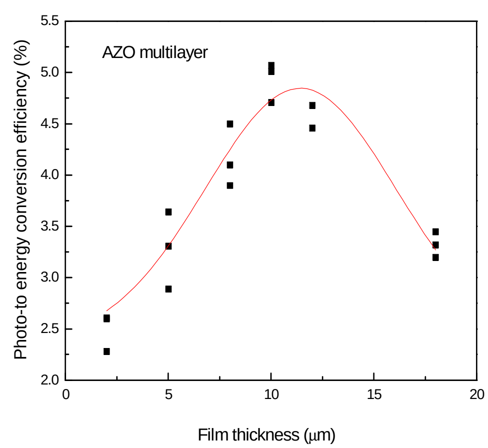

Figure 3 Variation of the light to energy conversion efficiencies of the DSSC with the thickness of the mesoporous TiO_2 layer grown on AAA

Here, with 4 cycle of coating, TiO_2 thickness of 8-12 μm in DSSC gives maximum efficiency. Other research group also use 10μm thick TiO_2 for the fabrication of DSSC which gives average optimum efficiency [14]. This may be due to different type of TCO and TiO_2 deposition process.

Figure 4 presents the I-V characteristics of the DSSC fabricated on AAA, ITO and FTO. The I-V graph indicates that current density and voltage in AAA is higher than ITO. There is also improvement in conversion efficiency of DSSC in AAA than that of ITO and FTO. Generally, the changes in the conversion efficiency depends not only on the differences in the conductivities of the transparent layers but on the antireflection of the multilayers and the transmittance of the incident light. Suitable thickness of each layer in the AAA nanocrystalline TiO_2/dye stack with proper deposition will suppress the internal reflection and achieves a high photocurrent which increases the conversion efficiency of the DSSC. This can be achieved using continuous structure of deposited TiO_2 /AAA film. DSSC prepared using thick mesoporous TiO_2 electrode on the FTO and ITO have efficiency of 4.81 and 3.67 % respectively under 1 sun light.

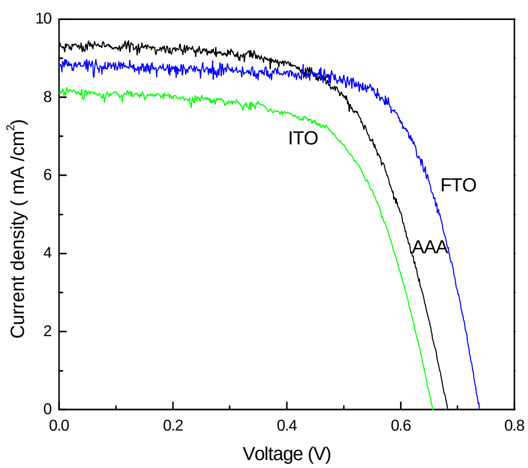

Figure 4. The current (I)- Voltage(V) graph of the DSSC fabricated with AAA, ITO and FTO glass.

It is reported that incorporation of a small quantity of ZnO into TiO_2 matrix increases th short circuit photocurrent and open voltage in the cell relative to that of bare TiO_2 film [3]. Based on this concept, ZnO incorporated TiO_2 DSSC was also fabricated. Figure 5 shows the I-V characteristics of DSSC having both TiO_2 and ZnO/TiO_2 electrodes. For ZnO/TiO_2 film, ZnO deposition was performed for 5 min using CVD process. It is observed that Jsc and Voc value increased using ZnO on the surface of TiO_2 film electrode. But, the F. F (fill factor) of the fabricated DSSC using ZnO incorporated TiO_2 was 0.65 which is marginally decreased from 0.69 of bare TiO_2 on AAA. Therefore, the overall efficiency of the DSSC increased to 5.54 % from 5. 07 % in case of AAA based cells. The increase of Jsc in case of cell containing ZnO/TiO_2 is due to enhanced light scattering by the ZnO/TiO_2 film relative to that by the TiO_2 film. It is also observed that, the bare TiO_2 film shows less absorbance than that of the ZnO/TiO_2. Generally, the free electron density in the TiO_2 conduction band indicates the absorbance [17]. The higher value of Jsc in DSSC is directly related to the higher free electron density which shows by higher absorbance of the ZnO/TiO_2 film [18]. The DSSC using developed low cost TCO and low-cost mercurochrome dye is a new step. Details study on the optimization of the film preparation along with the merits of such film is further necessary for the development of highly efficient DSSC.

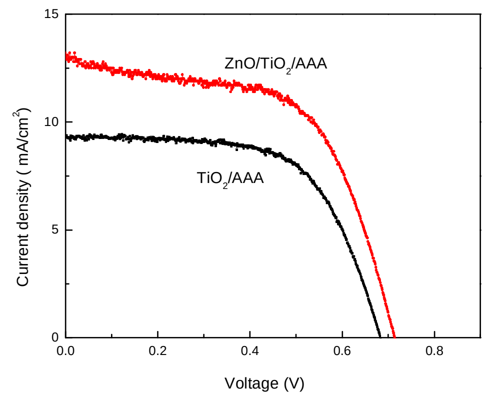

Figure 5. The I-V characteristics of DSSC with bare TiO_2 and ZnO/TiO_2 film electrodes.

4. Conclusion

Suitable multilayer TCO electrode with sheet resistance of 7 Ω/sq and transmittance of more than 85 % are synthesized for fabrication of DSSC. The DSSC on AAA covered with ZnO/TiO_2 film yielded an overall cell efficiency of 5.45 %. This developed multilayer TCO can be used as transparent electrode for making improved DSSC or plastic DSSC.

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