Fabrication and Characterization of CuO-based Solar Cells

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Abstract

Cuprous oxide (CuO)-based solar cells with fullerene (C_{60}) were fabricated on indium tin oxide (ITO) by a spin-coating method. The microstructure and cell performance of the solar cells with the CuO/C_{60} structure was investigated. A photovoltaic device based on an $ITO/CuO/C_{60}$ hererojunction structure fabricated by the spin-coating method provided short-circuit current density of 0.18 $mAcm^{-2}$ and open circuit voltage of 0.04 V under illumination. The crystal structure of the CuO active layer was examined by using X-ray diffraction. The energy levels of the present solar cells are also discussed.

Keywords: Solar cell, Copper oxide, Photovoltaic property, Optical property

1. Introduction

Solar cell technology for future energy resources has been progressed recently. Silicon is used as the semiconductor material for conventional solar cells, and the cost reduction of the solar cells is one of the most important issues. Cu oxides such as CuO and Cu_2O are one of the candidate materials. The features of copper oxide semiconductors are high optical absorption and nontoxic and low cost fabrication (Olsen, Bohara & Urie, 1997; Herion, Niekisch & Schari, 1980). CuO and Cu_2O are p-type semiconductors with band gaps of ~1.5 eV and ~2.0 eV, respectively, which are close to the ideal energy gap for solar cells and allows for good solar spectral absorption due to these direct band gap. The highest efficiency of ~2% for Cu_2O solar cells has been obtained by using the high-temperature annealing method and an expensive vacuum evaporation technique (Mittiga et al., 2006). Effcient hetero-junction solar cells with p-Cu_2O and n-ZnO fabricated by electro-deposition and photochemical deposition methods have been investigated and reported (Izaki et al., 2006; Izaki et al., 2007).

The purpose of the present work is to fabricate and characterize solar cells with CuO/C_{60} structures. Fullerene (C_{60}) is a good electron acceptor, and has been used as the n-type semiconductor active layer for organic cells (Jeong et al., 2007; Takeda et al., 2009).

2. Experimental procedures

3.6 g of copper acetate and 30 g of oleylamine were introduced in 70 g of 1-heptanol. The solution was heated at 100 °C for 2 hours. Then, a hot 1-heptanol solution containing 1.9 g of NaOH was quickly added. After 2h of reaction, the solution was cooled down to 25 °C. Throughout the process, the reaction system was subjected to mechanical stirring and purged with N_2. At the end of the reaction, the particles were collected by centrifugation and washed several times with a mixture of toluene and methanol. Finally, CuO particles were dispersed in toluene.

A thin layer of polyethylenedioxythiophen doped with polystyrene-sulfonic acid (PEDOT:PSS) (Sigma Aldrich) was spin-coated at 2000 rpm on pre-cleaned indium tin oxide (ITO) glass plates (Geomatec Co., Ltd., ~10 Ω/□). After annealing at 100 °C for 10 min in N_2 atmosphere, CuO layer was spin-coated at 1000 rpm. After annealing at 100 °C for 30 min in N_2 atmosphere, C_{60} layer was spin-coated at 1000 rpm. After annealing at 100 °C for 30 min in N_2 atmosphere, aluminium (Al) metal contacts with a thickness of ~100 nm were deposited as top electrodes by using vacuum evaporation, and annealed at 140 °C for 20 min in N_2 atmosphere. The solar cells were fabricated as a stacking structure of $ITO/PEDOT:PSS/CuO/C_{60}/Al$. Schematic illustration of the present solar cells is shown in Fig. 1.

The current density-voltage (J-V) characteristics (Hokuto Denko Corp., HSV-100) of the solar cells were measured both in the dark and under illumination at 100 mW/cm^2 by using an AM 1.5 solar simulator (San-ei Electric, XES-301S) in N_2 atmosphere. The solar cells were illuminated through the side of the ITO substrates, and the illuminated area measured 0.16 cm^2. Optical absorption of the solar cells was investigated by means of UV-visible spectroscopy (Hitachi, Ltd., U-4100). The crystal structures, phases and crystallite sizes of the copper oxide and C_{60} thin film were investigated by wide X-ray diffractometer (XRD, PHILIPS X'Pert-MPD System) with CuKα radiation operating at 40 kV and 40 mA.

3. Results and discussion

A solar cell with a CuO/C_{60} structure provided power conversion efficiency (η) of 1.8×10^{-6} %, fill factor (FF) of 0.25, short-circuit current density (J_{sc}) of 0.18×10^{-3} mAcm^{-2} and open-circuit voltage (V_{oc}) of 0.04 V. The photocurrent was observed under illumination and the CuO/C_{60} structure showed characteristic curves with short-circuit current and open-circuit voltage. Figure 2 shows measured optical absorption of thin films. The heterojunction and bulk heterojunction solar cells were denoted as CuO/C_{60} and CuO:C_{60}, respectively. Individual CuO and C_{60} thin films were also measured. The CuO/C_{60} structure shows high optical absorption in the range of 400 nm and 800 nm.

Transmittance spectrum of 100 nm thick CuO film, deposited on ITO, is presented in Fig. 3(a). From this spectrum, the optical absorption coefficients (α) of this film was determined from the spectral transmittance using the next equation, $α = 1/d \cdot \ln(1/T)$. Where d is the film thickness and T is the transmittance (Jhin-Wei Chen et al., 2011). For determination of the optical band gap energy (E_g), the method based on the relation of $αhν = A(hν-E_g)^{n/2}$ was used, where n is a number that depends on the nature of the transition. In this case its value was found to be 1 (which corresponds to direct band to band transition).

Figure 3(b) is a Tauc plot, which shows $(αhν)^2$ versus hν for the CuO film. The intersection of the straight line with the hν-axis determines the optical band gap energy E_g (Georgieva & Ristov, 2002). It was found to be about 3.7 eV which is higher than the ideal band gap of the CuO crystal. Because of the large band gap energy, the short-circuit current density would be low.

All the crystalline components in the CuO and C_{60} thin films were investigated by XRD, as shown in Fig. 4. Diffraction peaks corresponding to CuO and C_{60} are observed in thin films, which consisted of cupric phase with monoclinic system (space group of C2/c and lattice parameter of a=0.4653 nm, b=0.3410 nm, c=0.5018 nm, β=99.481 °). The particle size was estimated using Scherrer's equation: $D = 0.9λ/B\cosθ$, where λ, B, and θ represent the wavelength of the X-ray source, the full width at half maximum (FWHM), and the Bragg angle, respectively (Oku et al., 2010). The crystallite sizes of CuO and C_{60} were determined to be 3.4 nm and 51.9 nm, respectively. To increase the efficiency of the CuO/C_{60} solar cell, small grain size of C_{60} and higher crystallinity of CuO would be necessary.

Energy level diagram of the CuO/C_{60} solar cell is summarized as shown in Fig. 5. Previously reported values were used for the energy levels (Oku et al., 2010; Oku et al., 2011). It has been reported that V_{oc} is nearly proportional to the band gap of the semiconductors, and control of the energy level is important to increase efficiency (Park et al., 2010). Compared to silicon with an indirect transition band structure, CuO with a direct transition band structure is more suitable for the optical absorption property. In addition, the ultrathin film of the CuO layers could provide efficient charge injection because of the high optical absorption. Although ZnO has been mainly used as an n-type oxide semiconductor for solar cells (Motoyoshi et al., 2010), C_{60} was applied instead of ZnO in the present work.

The advantage of the present C_{60} is a good acceptor material for solar cells and the simple film formation using a spin-coating method. Compared to previously reported CuO-based heterojunction solar cells prepared by a spin-coating method without a vacuum system were investigated in the present work. The low conversion efficiency of the present solar cells would be due to presence of CuO nanoparticle aggregation in the active layer.

4. Conclusions

ITO/PEDOT: PSS/CuO/C_{60}/Al solar cells were produced and characterized, which provided η of 1.8×10^{-6} %, FF of 0.25, J_{sc} of 0.18×10^{-3} mAcm^{-2} and V_{oc} of 0.04 V. The CuO/C_{60} structure showed high optical absorption in the range of 400 nm and 800 nm, and the E_g of CuO was found to be ~3.7 eV from the Tauc plot, which is higher than the ideal band gap of the CuO crystal and the short-circuit current density would be decreased. A crystallite size of CuO was determined to be 3.4 nm, and higher crystallinity of CuO would increase the efficiency of the CuO/C_{60} solar cells. The energy level of the present solar cell was proposed, and separated holes could transfer from the valence band of the CuO to the ITO, and separated electrons could transfer from the conduction band of

the CuO to the Al electrode, respectively. Formation of the CuO/C_{60} active layer with homogeneous distributed CuO nanopaticles would improve the efficiencies of the solar cells.

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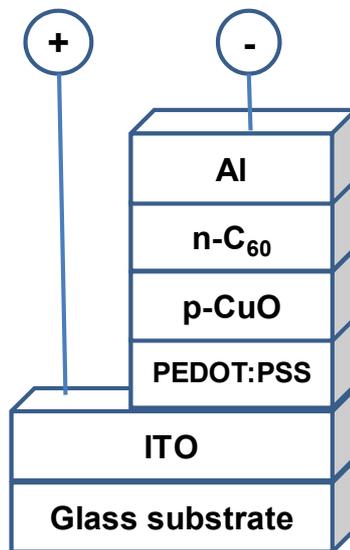
Figure 1. Schematic structure of CuO: C_{60} heterojunction solar cells
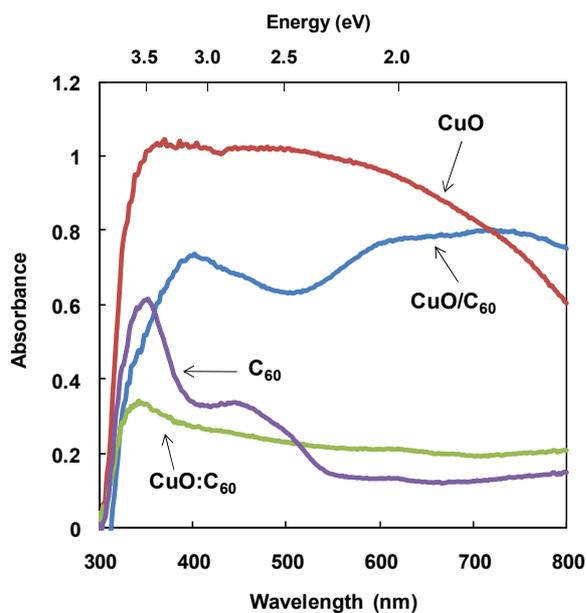
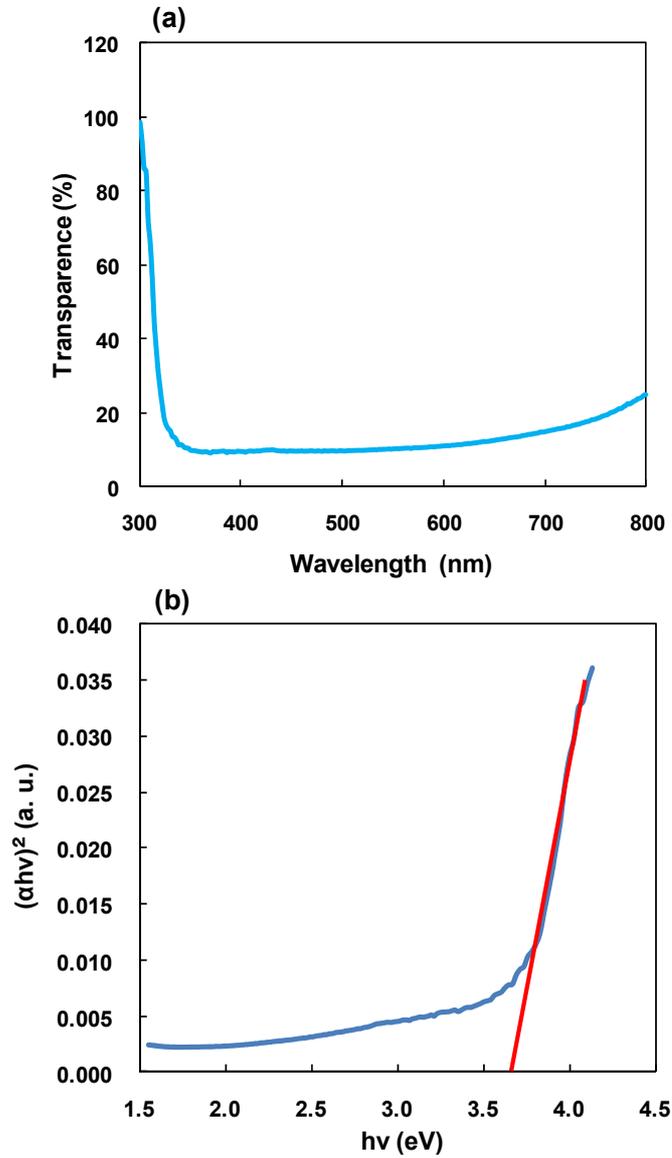

Figure 2. UV-visible absorption spectra of thin films

Figure 3. (a) Optical transmission spectrum of 100 nm thick CuO film, and (b) Tauc plot for the CuO thin film
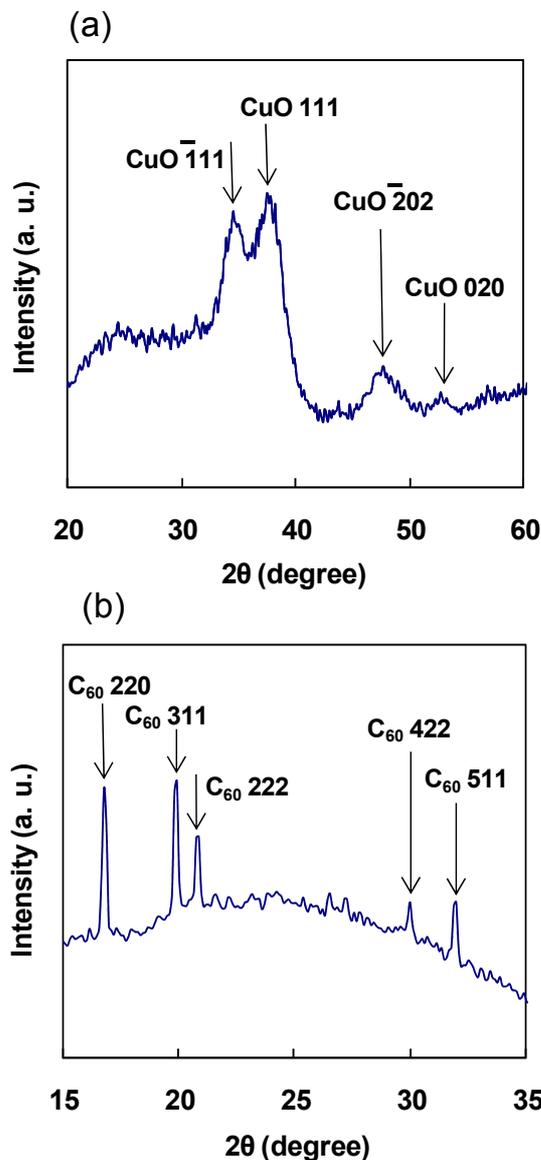
(a)

(b)

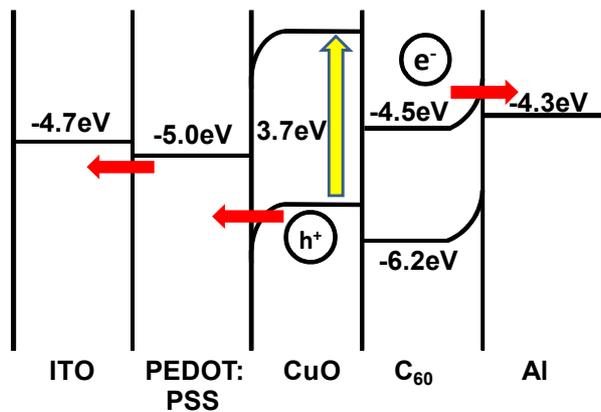Figure 4. X-ray diffraction pattern of (a) CuO and (b) C_{60} thin films

Figure 5. Energy level diagram of ITO/PEDOT: PSS/CuO/C_{60}/Al solar cells