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The effect of aging on the performance of open-air spin coated ZnO-CuO bulk heterojunction thin film solar cells

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The effect of aging on the photovoltaic characteristics of homely fabricated ZnO-CuO based bulk heterojunction solar cells (BHJSCs) including optical, electrical, chemical, and surface morphological properties of the spin-coated ZnO-CuO bulk thin films has been investigated in this article. The experimental results of open-air spin-coated bulk films on glass substrate reveal that the surface roughness, percentage of oxide composition, and electrical resistivity including the optical absorption is increased with aging. Also, the photovoltaic parameters of FTO/ZnO-CuO/Al BHJSCs have been decreased while the open-circuit voltage are increased noticeably with aging. These results indicate that the aging of bulk ZnO-CuO thin films has unavoidable consequences on the performance of ZnO-CuO-based BHJSCs.

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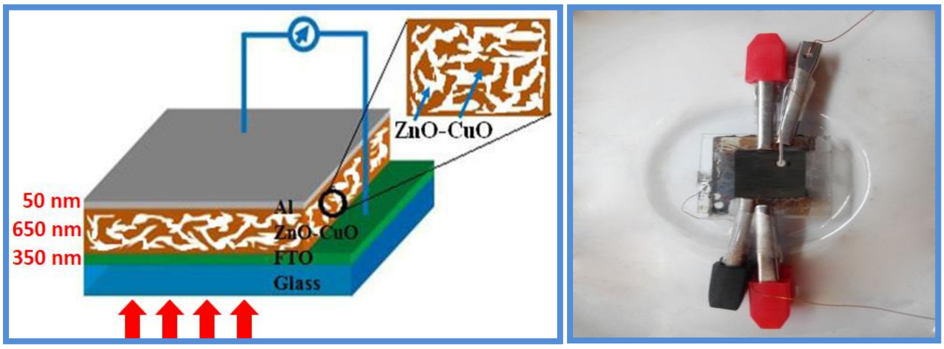
1. Introduction

Energy from fossil fuel resources like coal, petroleum, natural gas, and nuclear energy is becoming insufficient regarding rapidly increasing worldwide energy necessitates. The generation of electricity with renewable energy sources that are less polluting and economical is a way to mitigate such a huge amount of energy demands for the future. Solar energy harvesting is the most promising way for the production of renewable energy. Today, metal oxide heterojunction solar cell is considered as one of the most hopeful candidates for energy production. The solar cells fabricated using copper oxide (CuO) and zinc oxide (ZnO) are a potential alternative to conventional expensive silicon cells for some of the special properties like relatively low cost, naturally abundant, non-toxic, and environmentally benign whose theoretical efficiency of ~16 % [1–4]. ZnO is an important semiconductor material with excellent chemical and thermal stability at room temperature. Thin films of ZnO are important materials for semiconductor technology because of their versatile applications such as transparent conductors [5], solar cell windows [6], gas sensors [7], optical waveguides [8], photovoltaic devices [9], non-volatile memory [10], PH sensors [11], varistors [12] and surface acoustic wave devices [13-14]. On the other hand, CuO films are monoclinic crystal structure semiconductor with a direct bandgap of 1.2 - 2.1 eV. CuO has been used widely in the fabrication of solar cells, gas sensors, and cathodes in lithium primary cells, electronic devices, and electrochromic devices. [15]. Despite these advantages, the efficiency of ZnO-CuO heterojunction solar cell in practice remains low, achieving 4.12% with Cu_2O sheet fabricated by thermal oxidation (TO) at 1010 °C and only 1.43% with Cu_2O thin film via electrochemical deposition (ECD) at 65 °C, respectively [16,17]. Poor light-harvesting capability and low carrier collection efficiency are two of the most common issues in such ZnO-Cu2O heterojunctions [18-20].

There are many techniques for example thermal evaporation [21, 22], spray pyrolysis [23, 24], chemical vapor deposition [25-27], vacuum evaporation [28], magnetron sputtering [29, 30], and sol-gel spin coating deposition [31-34] are available for the preparation of ZnO-CuO bulk thin-film solar cells. Among these, the sol-gel spin coating method is of growing interest due to its ability to fabricate thin films of a large area with low cost and easy control of the microstructure of the films. Although the sol-gel spin coating method is a relatively simple technique compared with other film deposition techniques, there are still many factors affecting the quality of ZnO/CuO thin films [35-42]. Though there are some articles already that have been reported based on the effects of ZnO or CuO sol concentration and aging, pre-heating and annealing treatment on ZnO/CuO thin films have been reported but the aging effect on different properties of ZnO–CuO bulk heterojunction solar cell is seldom studied.

This contribution aims to investigate the influence of aging on the output parameters of homely fabricated FTO/ZnO-CuO/Al bulk heterojunction solar cells; open-circuit voltage Voc, short circuit current density Jsc, fill factor FF and therefore the efficiency η. Additionally, the aging effect on other fundamental properties of spin-coated ZnO-CuO bulk thin films like surface morphology, structural, optical, and electrical has been investigated in detail.



(a) *(b)*

Fig. 1. (a) Schematic diagram and (b) Top view of spin-coated ZnO-CuO based bulk heterojunction solar cell (color online)

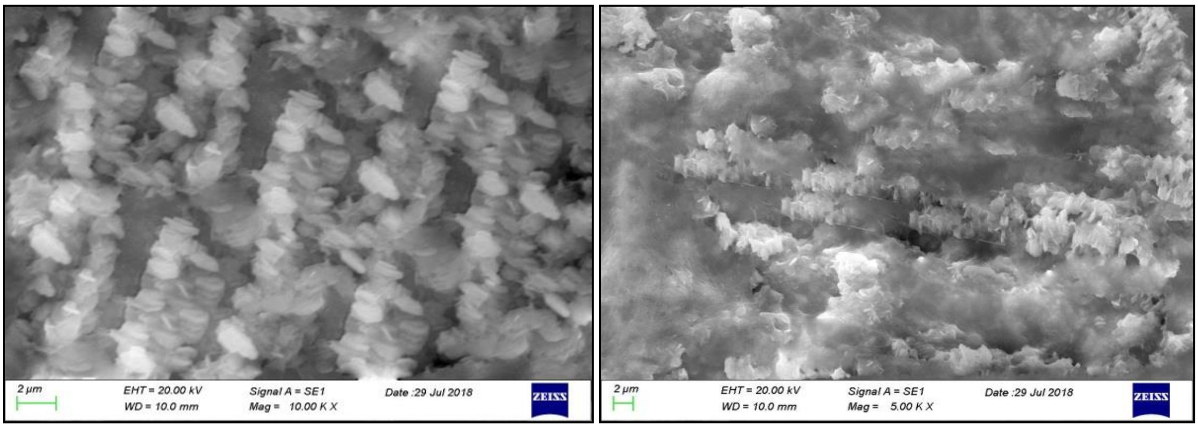
2. Materials and methods

The zinc acetate dihydrate [$Zn(CH_3COO)_2.2H_2O$] and copper (II) acetate monohydrate [$Cu(CH_3COO)_2.H_2O$] were used without any purification as the precursor of ZnO and CuO were collected from Merck Specialties Pvt. Ltd. and Loba chemic. Ltd. respectively. The bulk solution was prepared by the use of a single pot synthesis process. First, each of the precursors was measured separately: 2.19 g of zinc acetate and 1.99 g of copper (II) acetate are dissolved in 20 mL double distilled water in two separate beakers to prepare of 0.2 M bulk solution, stirred the solutions 10 minutes, then transferred both solutions (40 mL with 1:1 ratio) in a single beaker. This bulk solution was stirred vigorously at the temperature of 60 °C for 30 minutes. After that, this prepared bulk solution was deposited on FTO coated (350 nm) glass substrates (2x2 cm) using a simple spin-coating technique (with 500 rpm for 10s (warm-up speed and time), 2000 rpm for the 30s) and this process was repeated to make desired film thickness. In this study, the ZnO-CuO bulk solution was coated 5 times (~ 650 nm, the measure by Dektak Veeco 150 Profilometer) to ensure the suppression of the optical loss and improve the spectral absorption at wide ranges of the solar spectrum. The cleaning procedure of the FTO coated glass substrate was involved the following: mechanical washing in water with detergent, 2 times rinsing in DI water for removing the dust and particles additives and afterward, the ultrasonication in acetone, IPA and DI water each of 10 minutes respectively to remove the chemical residues and followed by the drying. Thereafter, the fabricated cells were annealed at 300 ºC for 30 minutes. The back-contact aluminum (Al) was deposited using a thermal evaporation technique with Edward-306. After the completion of the fabrication procedure, the fundamental characteristics of the as-prepared cell such as surface morphology, chemical, optical, electrical, and photovoltaic were studied. Afterward, this cell was preserved in the open air (at room temperature and pressure with 60%RH) for 60, 120, and 180 days. A similar study of the different characteristics as done for the as-prepared cell was performed in each of the aging stages. The photovoltaic performances of FTO/ZnO-CuO/Al heterojunction bulk thin-film solar cells were measured using a solar simulator (AM1.5 illumination) with Keithley-2400 Source Meter. The fabricated bulk cell (top view) is shown in Fig.1 with its schematic diagram.

3. Results and discussion

3.1. Surface morphological study

Fig. 2 depicts the Scanning Electron Microscope (SEM) of spin-coated ZnO-CuO bulk thin films with different aging time (a) as prepared and (b) 120 days aged bulk film. It is seen from Fig.2a, as prepared ZnO-CuO bulk film has a relatively smooth, regular, and homogeneous surface with uniform grains. On the other hand, Fig.2b shows a comparatively non-uniform, uneven surface with cracks and holes including the enlarged grains for 120 days aged bulk film. This is due to the formation of the higher degree of ZnO-CuO composite, zonal densification of metal oxides as seen in SEM study as well as the thickness study at different positions over the aged film surface. For the as-prepared sample, the thickness has to be found of 650±5 nm (smooth and regular) measured at the different regions of the film surface whereas the thickness of 120 days aged bulk film varies (Δt~35nm) from 640-677nm. The adsorption of environmental constituents like H_2, O_2, CO, CO_2, H_2O, hydrocarbons, etc. by the bulk ZnO-CuO film surface accelerates the change of surface chemistry that causes such surface degradation. These environmental constituent adsorption phenomena by the surface of the oxide have been described in detail in the reported articles [43-44].

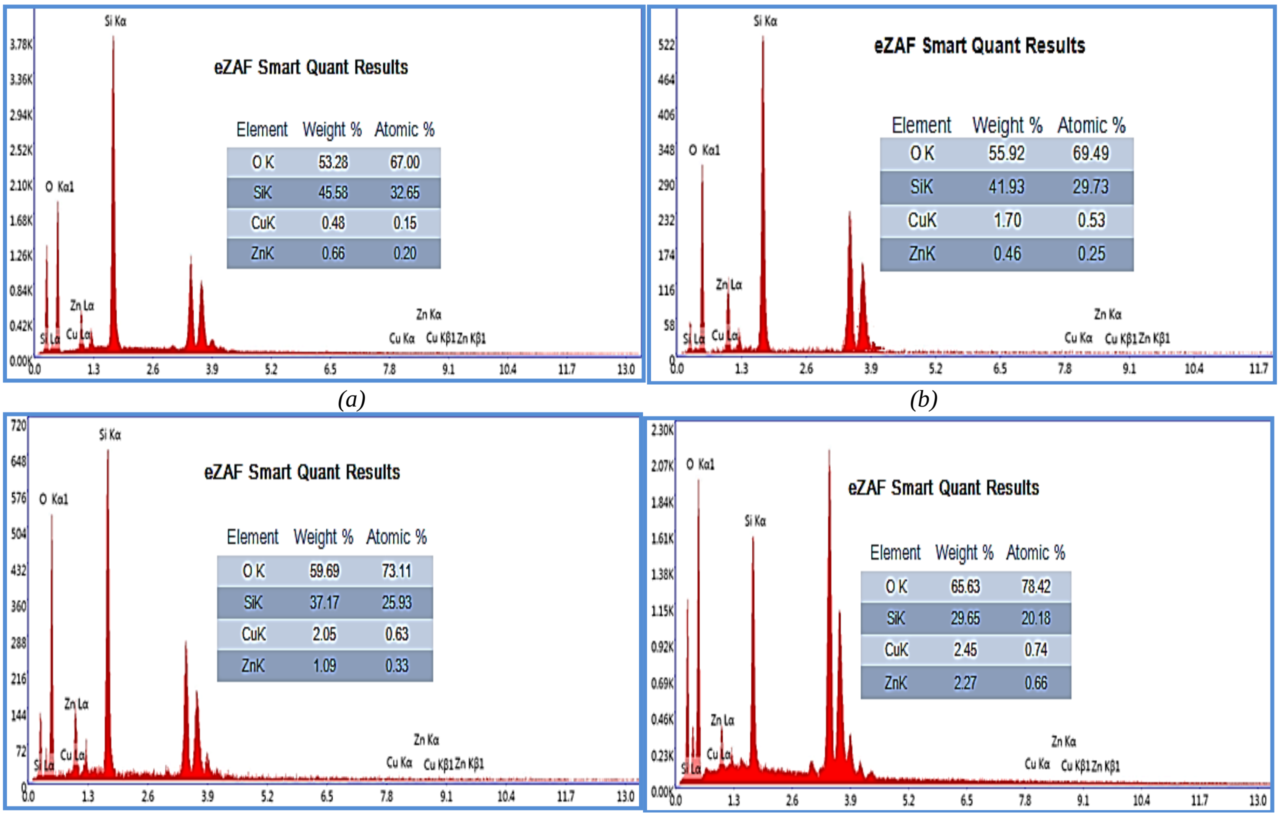


(a) *(b)*

Fig.2. Scanning Electron Microscope (SEM) of spin-coated ZnO-CuO bulk thin films (a) as prepared (b) after 120 days aging

3.2. Energy dispersive X-ray (EDX) study

Fig. 3 presents the results of the Energy Dispersive X-rays (EDX) of spin-coated ZnO-CuO bulk thin films (a) as prepared (b) after 60 (c) after 120 and (b) after 180 days of aging. It is found from Fig. 3, the atomic% of Si decrement from 32.65 to 20.18%. On the contrary, the atomic% of the Zn, Cu, and O is increased from 0.20 to 0.66%, 0.15 to 0.74%, and 67.00 to 78.42% respectively which indicates the increase of ZnO-CuO film thickness with aging. This may be due to the similar cause as observed in SEM, the formation of the higher degree of ZnO-CuO composite and possibility of the enlargement of grains size through the agglomeration of smaller grains including adsorption of the CO_2, O_2, H_2O, etc. by the bulk film surface that also revealed by the variation of the thickness of the bulk film surface with aging. In Fig. 3, the elements of carbon (C) and hydrogen (H) are absent due to the limitations of quantitative EDS analysis. Moreover, the experimental unavoidable confinement may another minor source of this increment of the Zn and Cu. Since the study of the bulk thin film was taken in different steps of aging time; there is a possibility of the selection of the measuring position on the bulk film surface as nearly similar but not the same position chosen during the previous steps of the EDS measurement though had tried best.



(c) *(d)*

Fig. 3. The Energy Dispersive X-rays (EDX) of ZnO-CuO spin-coated bulk thin films for (a) as prepared, (b) after 60 days, (c) 120 days, and (d) 180 days of aging (color online)

3.3. Optical study

Fig.4 shows the optical responses of spin-coated ZnO-CuO bulk thin films for as prepared, 60, 120, and 180 days of aging. It is seen from Fig.4a, as prepared ZnO-CuO spin-coated bulk film shows a relatively higher transmittance, and with the increase of aging time, this quantity of transmittance decreases. On the other hand, Fig.4b depicts the absorbance of the deposited ZnO-CuO thin films. The increment of the thickness due to the zonal densification and the development of the higher degree of ZnO-CuO composite with the adsorption of the environmental gasses; O_2, CO_2, H_2O, etc. from open-air, causes the decrement of transmittance and reversely the increment of absorption. The similar effect of the decrement of transmittance of individual ZnO films with aging and the adsorption phenomena of oxide films has been described in detail in the reported articles [35,43]. The average thickness of the bulk film was found to be of ~650 nm, ~666 nm, ~677 nm, and ~681 nm for as prepared, 60 days, 120 days, and 180 days of aging respectively selecting the same (nearly) position of the bulk film surface. Also, it is noted that the optical study has been performed for the nearly same position as the bulk film surface at different steps of the aging.

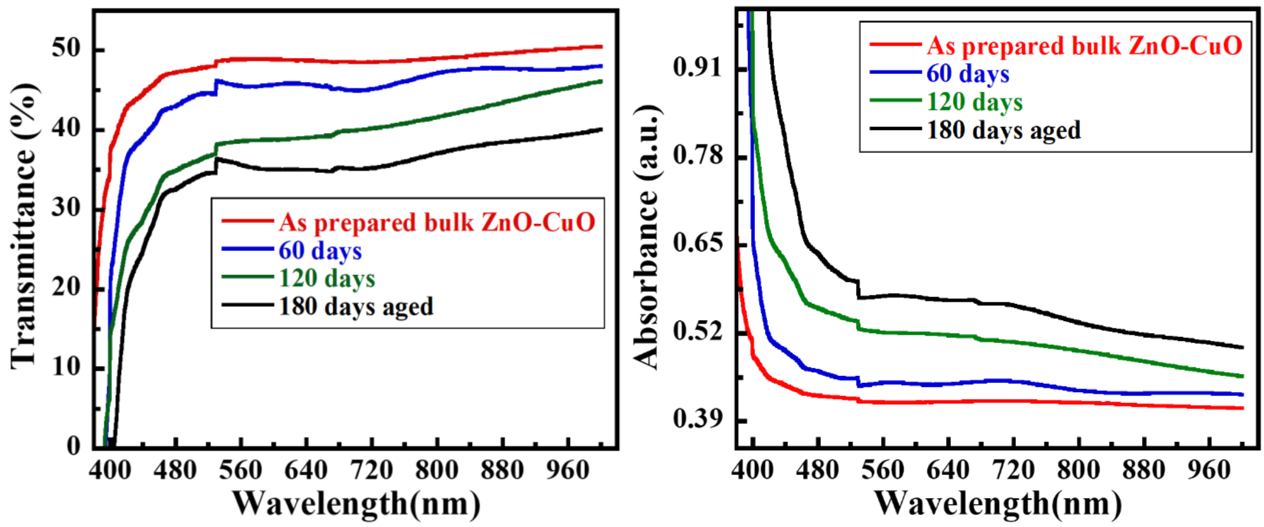


Fig. 4. Optical (a) transmittance and (b) absorbance of as prepared, 60, 120, and 180 days aged ZnO-CuO bulk thin films (color online)

3.4. Resistivity vs temperature study

Fig. 5 shows the electrical resistivity of spin-coated ZnO-CuO bulk films for as prepared, 60, 120, and 180 days of age at the temperature ranges from 30-350 ^{o}C. Conventionally, the electrical resistivity is greatly influenced by the temperatures. It is observed that the resistivity of the bulk films decreases dramatically (from ~4.55 to ~2.75 Ω-cm) up to 200 °C and decreases slowly afterward. This might be due to either an increase in the mobility and/or increase of the carriers with temperature. The increase of the temperature is attributed to larger grain sizes, thus leading to an increase of mobility as reported in the reported literature [45]. There are comparatively small numbers of free charge carriers for the case of room temperature but the electronically active carriers increase when the temperature increases, thereby leading to an excess carrier in the conduction band of the active metal oxides. This increase of active carriers due to thermal excitation giving rise to the conductivity of the bulk films conversely decreased resistivity. Besides, the increment of the resistivity is very small ($\Delta\rho \sim 0.25$ Ω-cm) with the aging of 60, 120, and 180 days up to the temperature of 50 ^{o}C where the increment of the resistivity $\Delta\rho \sim 0.5$ Ω-cm has been observed at the temperature of >50 ^{o}C. The development of zonal densification therefore voids and cracks that are revealed by the variation of the film thickness may the origin of this small increment of the resistivity of the bulk ZnO-CuO thin films with aging. The changing nature of the bulk ZnO-CuO film resistivity for different temperatures is much similar as found for Zn oxides thin film alone [46].

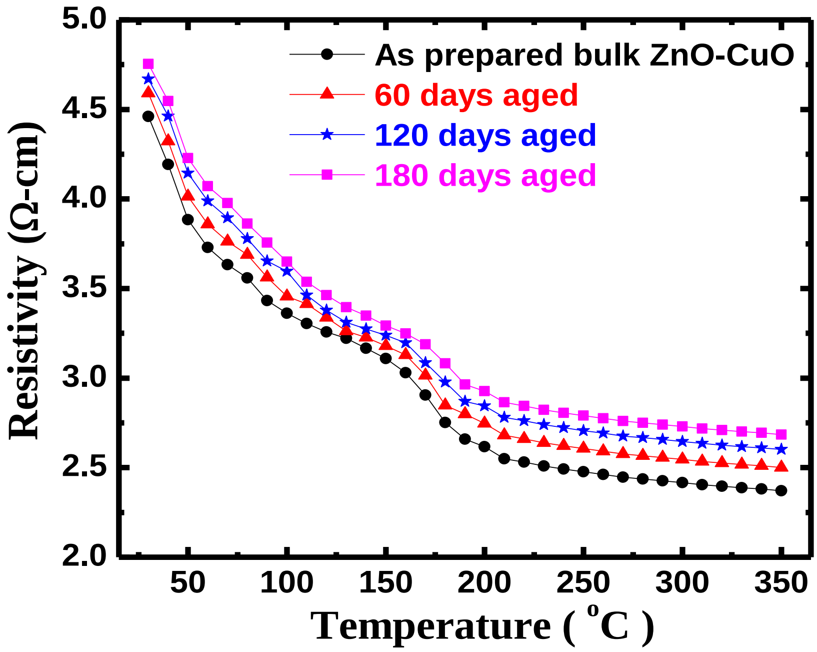


Fig. 5. The resistivity vs temperature of spin-coated ZnO-CuO bulk thin films for as prepared, 60, 120, and 180 days of aging (color online)

3.5. Photovoltaic study

Fig. 6 shows the J-V characteristics of open-air spin-coated bulk heterojunction solar cells having the hetero-structure of FTO/ZnO-CuO/Al for the various aging time of 0, 60, 120, and 180 days. It is observed from Fig.6, the as-prepared solar cell offers better performance than aged cells though the Voc sustains on an increasing trend. With the aging time, the FF and Jsc decrease (~5% and ~2.25 mA/cm^2 respectively) while the Voc increases ~0.7 V (from 0.5 to ~1.2 V). The "microstructure" of active materials is a crucial factor for changing the Voc of blend (mixed of both n and p-type semiconductors) heterojunction solar cells that comprises domain size, the existence of energetic barriers, electric field gradients, or dipoles at donor-acceptor interfaces of the bulk blend semiconductors, the columbic radius for the capture of charge carriers, the spatial distribution of intra-band trap states, etc. [47]. Therefore, the influence of the bulk film microstructure on the energy levels of the materials, charge carrier dynamics and energetic disorder is essential for understanding the change of the Voc. In this perspective, the bulk ZnO-CuO microstructure at the post of the cell fabrication and the changes of the microstructure dimension with aging affects the Voc noticeably like other factors: temperature, recombination, carrier density, charge transfer state, etc [48]. As seen from the SEM study, the bulk film densification has been observed with aging that may cause the improvements of the deposited bulk ZnO-CuO blend microstructure. This microstructural improvement offers adequate carrier lifetime and consequently, such increment of Voc as found in Fig. 6 has been observed. Moreover, articles on ZnO/CuO metal oxides deposited and characterized at different conditions have been reported from the same laboratory [49-54].

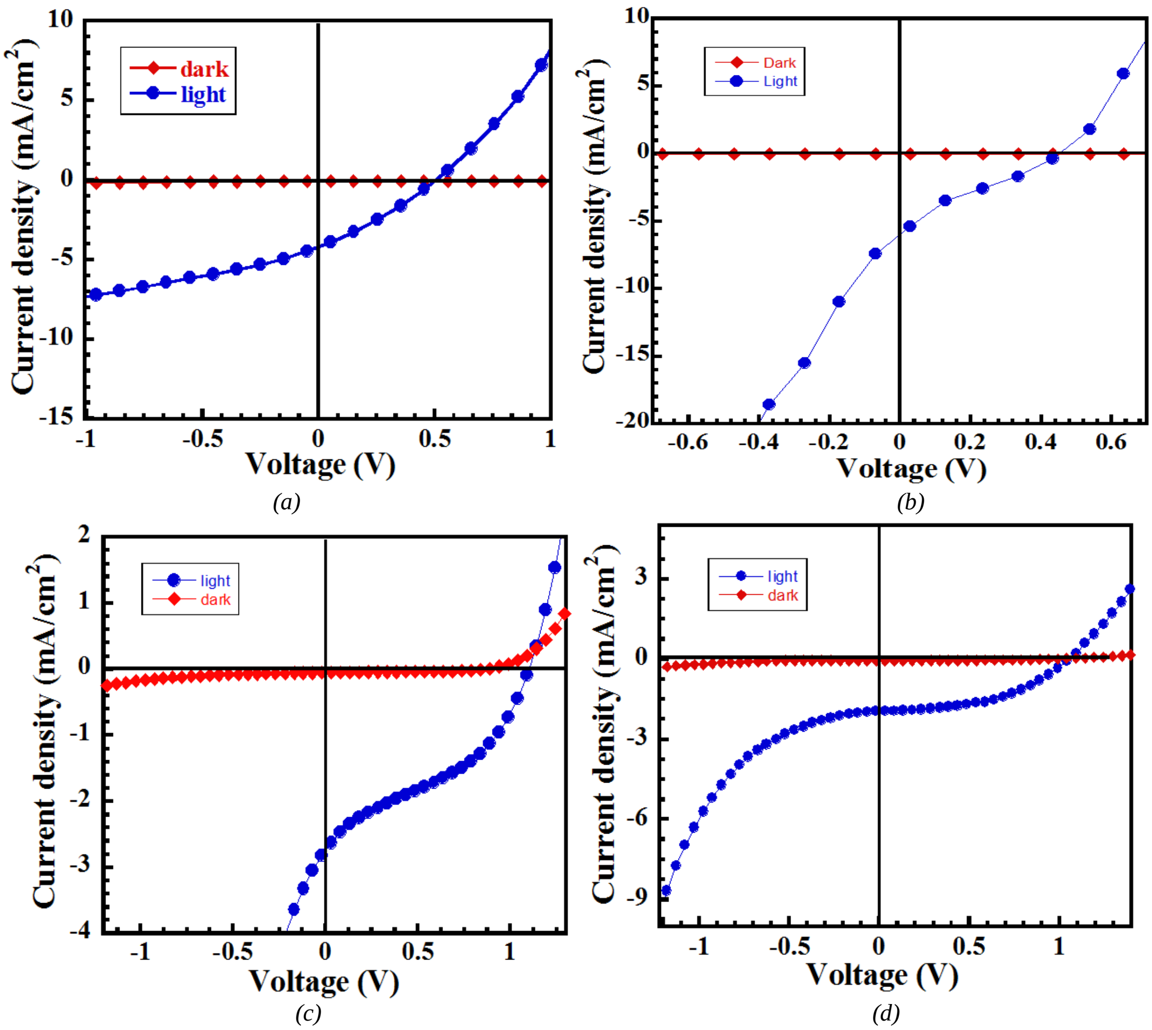


Fig. 6. J-V characteristics of ZnO-CuO spin-coated bulk thin films heterojunction solar cell (a) as prepared (b) after 60 days (c) after 120 days and (d) after 180 days of aging (color online)

Table 1. Photovoltaic parameters of ZnO-CuO spin-coated bulk heterojunction solar cell

Parameters	As prepared	After 60 days	After 120 days	After 180 days
Open circuit voltage, Voc (V)	0.53	0.46	1.1	1.2
Current density, Jsc (mA/cm2)	4.85	5.20	2.8	2.75
Fill factor, FF (%)	47	46	41	43
Efficiency, η (%)	1.20	1.10	1.26	1.42

4. Conclusions

Scanning electron microscopy (SEM) of open-air spin-coated bulk ZnO-CuO thin films delineate that with the aging, there are increased surface roughness, cracks, and holes thus the film surface quality has been degraded. Energy dispersive X-rays (EDX) results reveal that the percentage of oxygen (atomic and molar) increases noticeably with aging which refers gradually increased oxidation of the active materials due to adsorption. The optical transmittance decreases and conversely increases the absorbance of the spin-coated bulk thin films that are revealed by optical results. Electrical resistivity of the deposited films also increases though a small extent with aging as confirmed by the four-point probe method. The key characteristics like photoconversion efficiency η, short circuit current density Jsc and fill factor of open-air spin-coated FTO/ZnO-CuO/Al bulk heterojunction solar cells decreases noticeably excluding the open-circuit voltage Voc. This increment of open-circuit voltage Voc may be due to the improvements of the deposited bulk ZnO-CuO blend microstructure for the bulk film densification with aging that offer adequate carrier lifetime as well as the change of the surface chemistry due to the adsorption of plenty of O_2, CO_2, H_2O, etc. from the open environment. Experimental results indicate that the aging time affects the open-air deposited bulk thin film properties and photovoltaic performance of spin-coated FTO/ZnO-

CuO/Al heterojunction solar cells conspicuously. Finally, it may conclude that researchers should be concerned about the aging effect during the fabrication of metal oxides based bulk heterojunction solar cells.

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