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Research Article

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Comparative Study on Modified Alumino-thermic Processed Si NPs doped CuO-ZnO Spin Coated Heterojunction: Tandem and Bulk

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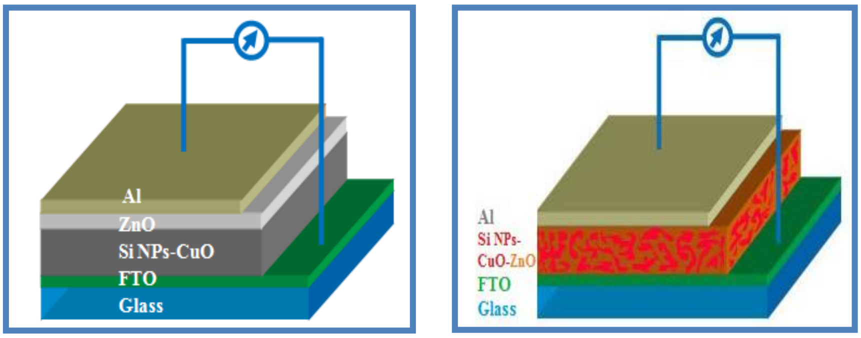
Abstract In this article, experimental investigation of structural, chemical and optical properties of silicon nano-particle (Si-NPs) doped copper oxide (CuO)/zinc oxide (ZnO) heterostructure thin films has been studied along with the photovoltaic characteristics of Si NPs doped one-pot synthesized CuO-ZnO based tandem and bulk heterojunction solar cells (HJSC). Si NPs were prepared first from silicon (Si) extracted from Padma river sand using modified Aluminothermic process through the mechano-chemical approach. Then, photoactive materials were doped with these Si NPs preserved in 2-propanol. Thereafter, SiNPs-CuO-ZnO thin films were prepared on a glass substrate and bulk and tandem heterojunction solar cells were fabricated onto FTO. EDX resultof Si NPs doped CuO-ZnO spin-coated bulk thin films confirms the presence of Si, Zn, O, and Cu in the film and formation of the junction is validated by quenching the PL peaks. Optical results reveal active materials with rich CuO-Si NPs offers better absorption than other ratios that support to enhance the efficiency of SiNPs -CuO-ZnOHJSC. Photovoltaic performances of spin-coated heterojunction solar cell refer, the tandem Al/SiNPs-CuO/ZnO/FTO HJSC offers better performance (more than η~0.14% larger) than bulk Al/Si NPs CuO-ZnO/FTO heterostructure solar cells. Above results indicate that CuO-ZnO based heterojunction solar cells are very promising with highlighting the spin coating with Si NPs as performance booster of tandem heterojunction thin-film solar cells.

Keywords ZnO-CuO, Si NPs, Heterojunction, Spin coating, thin films, Bulk and Tandem

1. Introduction

Due to rapidly increasing worldwide energy necessitates nonrenewable energy resources like coal, petroleum, natural gas, and nuclear energy is becoming insufficient. Generation of energy with less polluting as well as economical is a way to mitigate such a huge amount of energy demands for the future world. Photovoltaic cells (PVs) have attracted increasing attention as an effective and sustainable energy source. Choice of solar materials depends on various properties of associated active materials like band gap, absorption coefficient, diffusion length, thermodynamic compatibility and recombination velocity [1]. Silicon (Si) is highly efficient and core materials for its narrow band gap 1.1 eV having with above properties but a very expensive one. Today, metal oxide heterojunction solar cell is considered as one of the most optimistic, expectant and promising candidates for energy production. The solar cells fabricated using copper oxide (CuO) and zinc oxide (ZnO) are a prospective alternative to conventional expensive silicon cells for some of the special properties like naturally abundant, non-toxic, relatively low cost and environment benign whose theoretical efficiency of ~16% [2–6]. With excellent chemical and thermal stability ZnO with band gap of 3.3 eV is an important transparent window material for photovoltaic technology at room temperature because of its versatile applications such as transparent conductors [1], solar cell windows, gas sensors, pH sensors, optical waveguides, photovoltaic devices, non-volatile memory, varistors, surface acoustic wave devices[7-10] . Conversely, as absorber CuO films are monoclinic crystal structure semiconductor with a direct band gap of 1.2 - 2.1 eV. Moreover, the prospective application areas of copper oxide include the chemical industry, biosensors, Lithium-ion batteries, photo-catalysis, photoluminescence, gas sensors and optoelectronic and [11,12]. Literature reports several methods for the synthesis of CuO and ZnO such as thermal oxidation, potentiostatic deposition, activated reactive evaporation, spray pyrolysis, laser ablation, micro-emulsion, sonochemical, microwave and liquidphase synthesis [13,14]. Most research has been carried out by using n-ZnO/p-CuO hetero-junctions despite comparatively lower the cell efficiency of 4.12% fabricated using expensive and up to 2% for low-cost technique [15]. Doping the photoactive materials with Si-NPs may play a highly promising result for the enhancement of absorption coefficient, therefore, the cell performance. In this study, Si NPs extracted from river sand following the reported articles [16-18] was doped in CuO first and then Si-NPs doped CuO-ZnO based thin films heterojunction solar cells with bulk and tandem heterostructure have been fabricated. Finally, different properties like structural, surface morphological, optical and photovoltaic are studied carefully.



(a) (b)

Figure 1: Schematic diagram of (a) tandem and (b) bulk heterojunction solar cells

2. Materials and Methods

Chemicals zinc acetate dehydrate [Zn $(CH_3COO)_2.2H_2O$],sodium hydroxide (NaOH) were purchased from commercial suppliers Merck Specialties Pvt. Ltd. and copper (II) acetate monohydrate [Cu$(CH3COO)_2.2H_2O$] from Loba chemic. Ltd and these were used without further purification. 1.99 g cupric acetate monohydrate and 2.19 g zinc acetate dehydrate were dissolved in 20 mL 2-propanol solvent simultaneously to prepare 0.2 M ZnO-CuO sol under vigorous magnetic stirring for 30 min. 2-propanol contains Si NPs prepared from Si extracted from sand using Aluminothermic reaction through ultra-sonication about 2 hours. Using synthesized Si NPs-ZnO-CuO thin film on the glass substrate and cells on FTO coated glass substrate were fabricated using spin coating technique at 500 rpm for 10s (warm-up speed and time) then 2000 rpm for the 30s. Thereafter, the fabricated cells were annealed at 300 ºC for 5 min. Back contact aluminum (Al) was deposited using a thermal evaporation technique with Edward-306. The schematic diagram and fabricated solar cell are shown in Fig.1.

3. Results and Discussion

A. Structural study: X-ray diffraction, SEM and FTIR study

Figure 2a is an XRD scan of a Si-NPs doped ZnO-CuO heterojunction thin film. The phase separation between ZnO and CuO is visible confirming the existence of ZnO-CuO and CuO and ZnO revealing the peaks at 2θ values of 31.8º, 36.3ºand 67.1º correspondingly with the structure of hexagonal wurtzite for ZnO and monoclinic for CuO semiconductor, these peaks are well-matched with reported articles [19-21]. Also, the prominent peaks at 28.3º for Si not only for the doping of Si-NPs in ZnO-CuO but also the substrate used to deposit the films. Fig. 2b depicts cross-sectional SEM images of spin-coated ZnO-CuO tandem heterojunction solar cell.

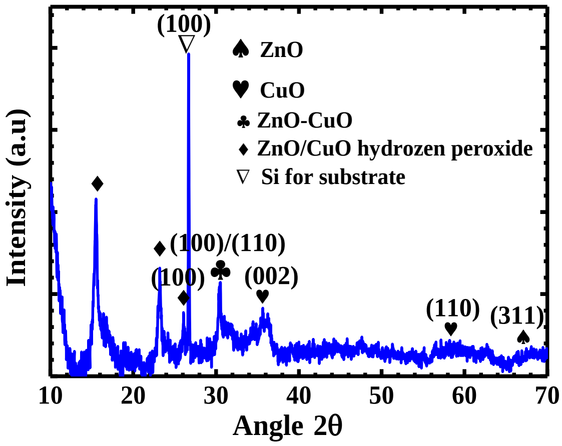


Figure 2a: XRD of spin-coated ZnO-CuO heterojunction thin film

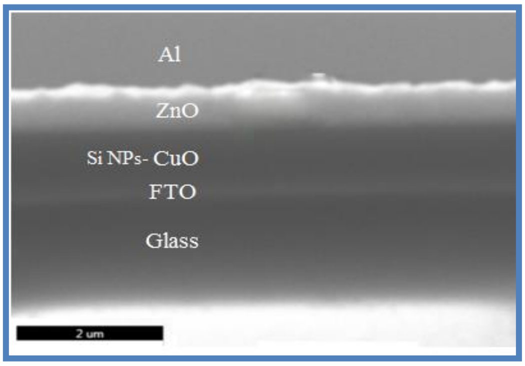


Figure 2b: SEM of spin-coated ZnO-CuO THJSC

Fig. 2b depicts cross-sectional SEM images of spin-coated ZnO-CuO tandem heterojunction solar cell. The film surface of spin-coated tandem heterojunction solar cell was found homogeneous, dense with fewer cracks and pin-holes.

In Fig. 3, FTIR peak at 644 cm^{-1} confirms that the synthesized product is CuO as reported [19-21]. 523 and 928 cm^{-1} reveals the bonds of ZnO that agree well with a stated article [22] and 1082 cm^{-1} refers to Si [26]. It is also observed a common peak in both samples at 1020 cm^{-1} that indicate the existence of C-OH which well matched with reported articles [27,28]. A peculiar FTIR peak for Si at 1082 cm^{-1} only seen in Si-NPs doped CuO-ZnO spin-coated thin film.

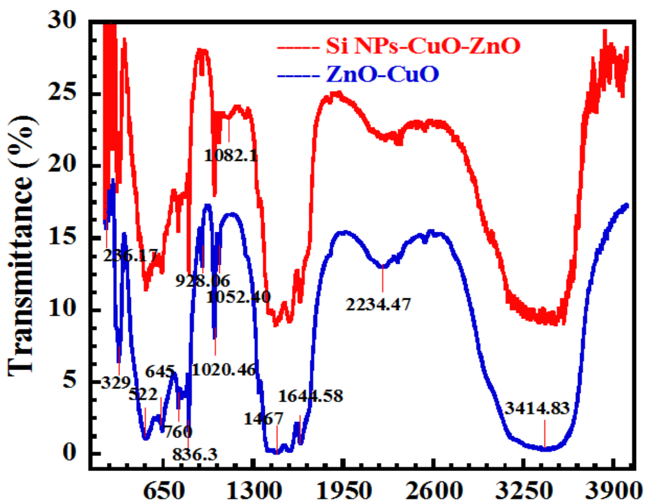


Figure 3: FTIR of spin-coated ZnO-CuO and Si NPs-CuO-ZnO

B. Energy dispersive X-ray (EDX) and PL study

Fig.4 presents the results of the Energy Dispersive X-rays (EDX) of spin-coated bulk SiNPs-CuO-ZnO bulk thin films. The dominated very high peak not only for Si NPS but also for glass substrate was used for deposition the films. This result confirms the elemental presence of Zn, O, S, and Cu in film.

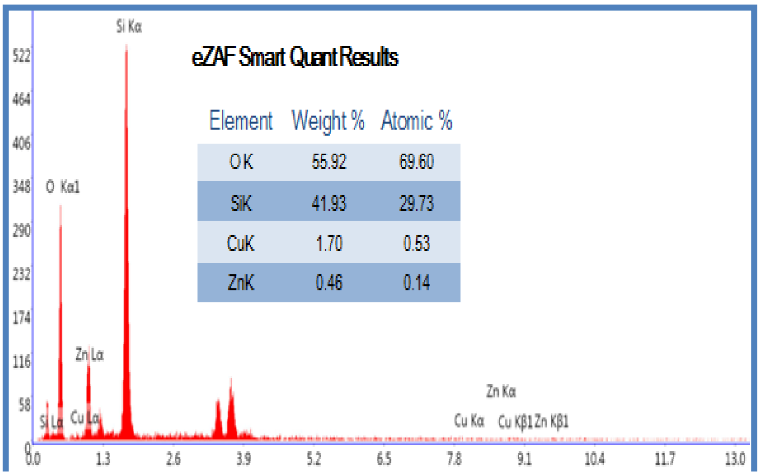


Figure 4: Energy Dispersive X-rays of Si-doped Spin-Coated CuO ZnO(2:1) thin films HJSC

Photoluminescence peak quenching shown in Fig. 5 confirms the formation of heterojunction between ZnO and CuO. This result may due to a charge transfer which occurs between the donor and the acceptor and for defect states which leads to non-radiative recombination between electron-hole pairs. Fig. 5 clearly shows the peak intensity of individual ZnO much higher than the PL peak of ZnO-CuO that indicate an energy transfer between donor and acceptor and thereafter the existence of the p-n junction in the deposited films.

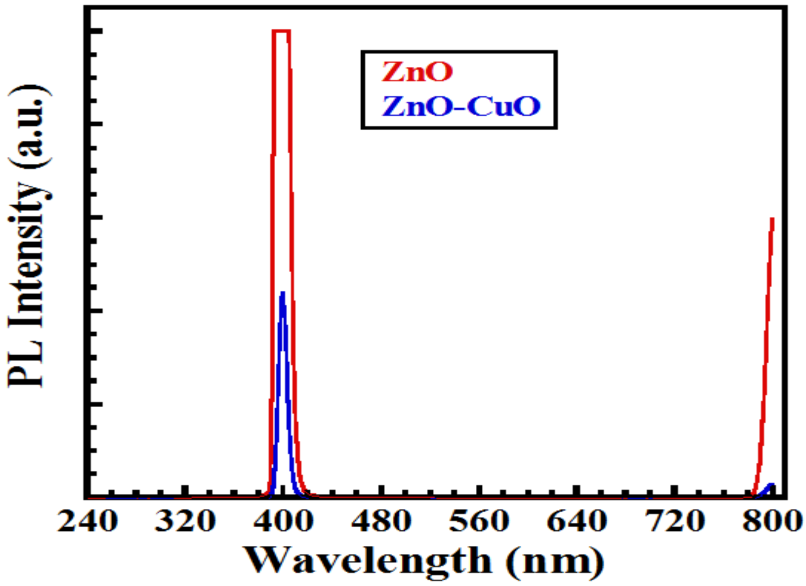
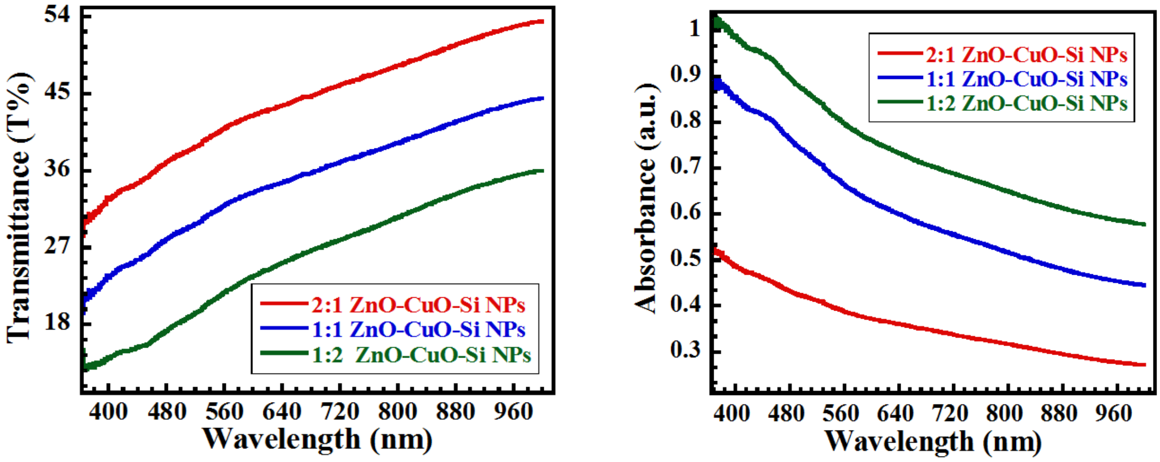


Figure 5: PL study of ZnO and bulk ZnO-CuO (PL peak quenching)

C. Optical study

Fig. 6 shows the optical response of spin-coated ZnO-CuO-Si NPs bulk thin films. Optical absorbance for three different ratios of Si NPs doped CuO and ZnO based thin films is shown in Fig.6.It is plainly seen from Fig. 6a, as prepared ZnO-CuO-Si NPs spin-coated bulk thin film with rich of CuO-Si NPs offers a relatively low amount of transmittance and conversely high absorbance shown in Fig. 6b.



(a) (b)

Figure 6: (a) Transmittance &(b) Absorbance of spin-coated Si NPs-CuO-ZnO bulk thin films

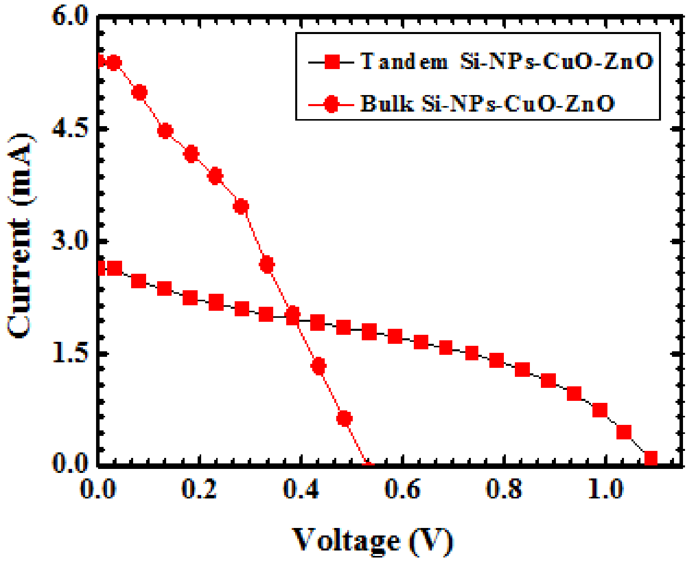


Figure 7: I-V characteristics of spin-coated Si NP-CuO-ZnO based heterojunction solar cells

D. Photovoltaic performance

Fig. 7 shows the J-V characteristics of spin-coated ZnO-CuO-Si NPs tandem and bulk heterojunction solar cell (cell are 1.44 cm^2). It clearly seen from Fig.7, the photovoltaic performance of the tandem heterojunction solar cell is comparatively larger than the performance of bulk HJSC though lower the current density.

Table 1: Photovoltaic parameters of spin coated Si NPs-CuO-ZnO solar cells.

Parameters	Bulk HJSC	Tandem HJSC
V_{oc} (V)	0.54	1.1
J_{sc} (mA/cm^2)	3.78	2.16
FF (%)	47	46

Table-1 presents the comparative study on the photovoltaic parameters open-circuit voltage $V_{oc,}$ short circuit current $J_{sc,}$ fill factor FF and efficiency η of bulk and tandem HJSC. Open circuit voltage in tandem HJSC higher, it may due to recombination of plenty of charge carrier for lengthening the diffusion length.

4. Conclusion

Scanning electron microscopy (SEM) image shows smooth, cracks and holes free with less roughness surface of open-air spin-coated heterojunction solar cell. EDX result confirms the elemental presence of Zn, Si, O, and Cu that supports FTIR result to confirm the existing bond of ZnO and CuO and Si. Optical results of bulk ZnO-

CuO-Si NPs refer that bulk thin films with rich CuO-Si NPs offer better absorbance than other ratios. And, the tandem structure of fabricated HJSC shows enhanced performance (η~1.10%)than bulk (η~0.96%) heterostructure. Experimental results indicate that tandem heterostructure with rich Si NPs-CuO absorber is the most hopeful for ZnO/CuO interface HJSC **highlighting the much promising spin coating technique for inexpensive and scalable photovoltaic fabrication.**

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