



Copper oxide based nanostructures for improved solar cell efficiency



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ABSTRACT

Resurgence of copper oxide based thin film solar cells demands exclusive methods of integrating various layers with superior constituents for increased solar–electric conversion efficiency. Exceedingly optically active nanostructured phase mixture of copper oxides was synthesized by an energy efficient hydrothermal process. Comprehensive structural and optical studies of these nanostructured copper oxides reveal its efficacy as a unique solar cell material. Excellent solar cell characteristics have been observed when these nanopowders are integrated with ZnO/CuO based thin films. X-ray diffraction, Raman micro-scattering, scanning electron microscopy, energy dispersive X-ray spectroscopy, UV–vis spectroscopy, atomic force microscopy, and optoelectronic measurements were employed to characterize these unified electronic devices. Solar cell measurements indicate a considerable increase in short circuit current density (J_{sc}) and open circuit voltage (V_{oc}) in the fabricated nanostructure powder–thin film hybrid solar cell devices. The solar cell efficiency of these nanopowder–thin film devices is found to be 2.88%. The physics behind this enrichment of solar cell properties has also been elucidated in the study. Exhaustive Raman spectroscopic and photoluminescence studies prove that multi-phonon scattering may play a major role for this enhancement. This integration of nanostructures with thin film solar cells can evolve to a new direction in photovoltaic technology.

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1. Introduction

Developing high-end solar cell devices by implementing inexpensive nanostructured materials and unique processing techniques is urgently required to sustain the ever demanding energy need. Copious new concepts for solar–electric energy conversion have been reported which challenge traditional photovoltaic (PV) devices based on the physics of semiconductor p–n junction diode [1]. Semiconductor materials used in solar cell technology are predominantly governed by their energy band gap, optical properties, and charge carrier diffusion length [2,3]. Highly efficient solar cell design necessitates optically active photovoltaic layers to enable nearly complete light absorption thereby increasing the rate of electron–hole pair generation. Considerable efforts have been made to develop state-of-the art nanostructured materials that absorb better at long wavelengths, so far with little success [4,5]. An option to increase the absorption of visible solar spectrum is by increasing the film thickness to improve the optical density. However, this leads to detrimental effects of exceeding the electron diffusion length through the nanoparticle network [6]. The ever demanding increased solar cell efficiency and reduced material consumption urgently require a decrease in active cell region thickness while preserving high optical absorption.

Copper oxide based semi-conductors are widely studied as photovoltaic materials [7], owing to its abundance and suitable optical properties for solar cell applications. These materials provide a unique possibility to tune the optical and electronic properties from insulating to metallic conduction, from band gap energies of 2.1 eV to the infrared at 1.4 eV, i.e. right into the middle of the maximum efficiency for solar-cell applications. Cupric oxide (CuO) and cuprous oxide (Cu₂O) are being widely used as a p-type semiconductor for designing solar cells [8,9]. There are innumerable examples of copper oxide based PV devices reported in literature, often prepared by using low-cost, solution-based methods [10–12]. Predominantly, all of these devices are either bulk or thin-film bilayer cells, and these types of cells suffer from the fact that the optimal material length scales for optical absorption and carrier extraction are contrary to one another. The use of nanomaterials represents a general approach to reduce both cost and size thereby improving efficiency in photovoltaic cells [13,14]. The physical mechanism underlying high external quantum efficiencies for photoluminescence in low dimensional materials is mainly due to the quantum confinement of excitons in a nanometer-scale crystalline structure [15,16]. As photons are absorbed by the material and charge carriers are produced, the average diffusion time (τ_d) from the bulk of the material to the surface has been shown to follow Eq. (1):

$$\tau_d = \frac{r^2}{D\pi^2} \quad (1)$$

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where r is the grain radius and D is the diffusion coefficient of the material [2]. A decrease of grain radius reduces the average diffusion time for the charge carriers thereby reducing recombinations, which forms the backbone of an efficient solar cell. Several device structures have been proposed, including nanopillar arrays with axial and radial junctions [17–19], and nanopillar collectors embedded in absorbing thin films [20]. Stringent control over the morphology of the nanomaterial itself has been shown to change the optical properties of a material [21,22]. The morphology associated with both the particle network and the inter particle contact area also dictates solar activities [23,24]. Band gap engineering for better optical properties is being widely used for metal oxides by the process of doping [25], and mixed oxide synthesis [26]. The constant effort to synthesize nanostructures with well-defined geometrical shapes and organizing them as 2- and 3-dimensional assemblies have further expanded the possibility of developing new strategies for light energy conversion [27,28]. Recently, we have reported a considerable change in the optical properties with the change in phase percentage, morphology and size of room temperature stable copper oxides [29].

The relative band alignment of two materials is important to understand the charge transport at the interface. For an effective charge separation there should be a type II or type III band alignment [30]. For better comprehension of the charge transport in thin films the p-type material should have both valence and conduction bands higher than the n-type material. The charge separation for an increased solar cell efficiency demands faster diffusion time of the charge carriers [31]. Assuming radiative recombination at the interface and proper alignment of the valence band of p-type material with the conduction band of n-type material gives the theoretical value of the open circuit voltage (V_{oc}) [32,33]. Integration of zinc oxide with copper oxide forms a type II band alignment which favors a perfect solar cell assembly [34]. Surface textures of a thin film as well as the morphology of nanoparticles play a major role in charge transport. Structural distortion in thin films could impede solar cell efficiency. In this paper we report a unique way of fabricating copper oxide based solar cells by integrating the thin films grown by pulsed laser deposition (PLD) and nanomaterials through drop-casting. The incorporation of nanostructured copper oxide powder with high optical absorption onto glass/ITO/ZnO/CuO thin film stack enhances the performance of solar cells. Subsequent annealing of the device at low temperature boosts the solar cell characteristics.

2. Experimental details

All chemicals were analytic grade reagents and used without further purification. The experimental details were as follows: (0.15 M) $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (GFS Chemicals – Assay 98.0–102.0%) was dissolved in (20 ml) deionized HPLC water (GFS Chemicals – Resistivity at 25 °C is 18.0 M ohm cm) under constant stirring. 45% (w/w) of KOH (GFS Chemicals – Assay 45%) was added to the resulting aqueous solution until the pH reached 13. The solution was transferred into a Teflon lined stainless steel autoclaves, sealed and maintained at 80 °C for 15 h. In order to investigate the growth mechanism of copper oxides, 10 ml ethylene glycol (GFS chemicals – 99% pure) was added to the aqueous solution before the hydrothermal process was started. After the completion of the hydrothermal process, the solid products were centrifuged (LW Scientific centrifuge – Model E8) at a rotation speed of 1800 rpm for 30 min, washed three times with distilled water and ethyl alcohol (Fisher Scientific, anhydrous and denatured), respectively, to remove the ions possibly remaining in the final product, and finally dried at 100 °C in air. Thin films of ZnO and CuO were deposited on glass/ITO substrates by pulsed laser deposition (Excel Instrument, PLD-STD-18) using a ZnO and CuO target. A KrF excimer laser (Lambda Physik, COMPEX 201) with energy density of 2 J cm^{-2} , $\lambda = 248 \text{ nm}$, pulsed duration of 20 ns, was used at a pulse rate of 10 Hz for the deposition. Thin films were grown at a growth temperature of 300 °C and

oxygen pressure of 1×10^{-5} mbar in the chamber. The base pressure of the deposition chamber was 1.2×10^{-9} bar. The thicker ZnO PLD grown film deposition was followed by a thinner PLD grown CuO film. Typical film thicknesses were approximately 100 nm and 50 nm for ZnO and CuO respectively, measured ex-situ by a profilometer (Veeco, Dektak 150). The nanostructured phase mixture of copper oxide was dissolved in toluene and sonicated for 5 min. The solution is then drop-casted and annealed at 130 °C in O_2 on the glass/ITO/ZnO/CuO heterostructures. The sample was annealed for different annealing times (1 h and 2 h). Low temperature annealing was performed to retain the structural integrity of the nanostructured phase mixture of copper oxide. The temperature of 130 °C in O_2 was sufficient enough for physisorption of the copper oxide nanostructures onto glass/ITO/ZnO/CuO thin film. The drop-casted film thickness was tracked via optical absorption, and the process was repeated to obtain films of the desired thickness. DC sputtering technique was used to grow high quality top silver electrodes (500 μm diameter). The devices were characterized using X-ray diffraction (Bruker, D8 Discover) θ –2 θ scan with $\text{CuK}\alpha$ ($\lambda = 1.5405 \text{ \AA}$); Rietveld analysis was done using *DiffraC^{plus}* Topas software. The morphology of as-prepared samples were characterized by scanning electron microscopy (FEI Quanta 200S) and atomic force microscope (Digital Instruments, DI 3100). The vibrational phonon modes and photoluminescence studies of the samples were determined by Raman spectroscopy using a 532 nm green laser, and 325 nm UV laser (Horiba Labram Raman-PL). Absorption characteristics spectra were measured by Ocean Optics HR 4000 spectrometer in conjunction with Ocean Optics Spectra suite software. Solar measurements were done using AM 1.5 solar light source (Newport, 69907 light source) coupled with Labview 2012 software programming. The variable voltage across the solar cell device was applied using a voltage source (Keithley, 230 programmable voltage source), and the current was measured by an electrometer (Keithley, 617 programmable electrometer).

3. Results and discussion

3.1. Copper oxide nanostructures

Hydrothermally synthesized copper oxide nanostructure was characterized to determine its optical superiority as compared to other synthesized nanopowders. Fig. 1 elucidates the optical effectiveness of the as synthesized copper oxide nanostructures, and its dependency on structural (X-ray diffraction) and molecular vibrational modes (Raman spectroscopy). The observed XRD pattern (black line) along with that of calculated (red line) using Rietveld analysis and difference (blue line) profile for the powder sample are shown in Fig. 1(a). XRD data indicate that the mixture of copper oxides formed crystallizes in space group symmetry of C12/c1 (monoclinic) and Pn–3 m Z (cubic) for CuO and Cu_2O , respectively. The crystal structure was extracted by Rietveld analysis and is represented in the inset of Fig. 1(a). The calculated bond angles (a, b) and the bond length (c) of the copper oxide nanostructures are 95.74°, 78.93°, and 1.961 Å, respectively. The calculated goodness of fit is 1.07. The calculations indicate a least deviation from the standard bond angles and bond length [29]. A larger deviation in bond angles and bond lengths from the pure CuO results in weak or strained bonds. The energy of the weak bonds is higher and these bonds can easily break to form defects in the atomic network [35]. Band tail states represent the energy states of electrons that form the strained bonds in the crystal network [36]. Increase in disorder and strained bonds increases the width of the tail states. The wave functions of the tail and defect states are localized within the structure and therefore these states are also referred to as localized states [37]. The mobility of the charge carriers are least in those localized states [36,37]. Incorporation of these nanostructures for fabricating a solar cell can actually be useful due to minimal density of the above-mentioned tail states.

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