Role of a Solution-Processed V$_2$O$_5$ Hole Extracting Layer on the Performance of CuO-ZnO-Based Solar Cells

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1. INTRODUCTION

Solar energy is one of the largest, flexible, and technologically advanced, sustainable, low-carbon choices available to satisfy the increasing global electric power demand. Photovoltaic devices are the most extensively used solar technology to utilize solar power. Electricity from photovoltaics could fill around 20% of the market for global primary energy by 2050.\(^1\) Crystalline silicon (c-Si) is the most successful photovoltaic technology available, which occupies about 90% of the current world photovoltaic market share.\(^2\) The cost per efficiency of the silicon solar cells is the main concerning issue to the photovoltaic researcher community.\(^3\) This issue has driven the researchers toward finding an alternative earth-abundant absorber material for the fabrication of low-cost highly efficient solar cells.

Inorganic metal oxides (IMOs) are earth-abundant materials and can be synthesized using very simple and low-cost environment-friendly chemical routes. With band gaps of 1.34–1.7 eV and a high absorption coefficient, CuO, a p-type IMO, could be one of the promising candidates for photovoltaic applications.\(^4,5\) CuO has some other excellent features like high minority carrier diffusion length,\(^6\) tunable electrical properties,\(^7\) etc. These features of CuO have enabled its use in the sensor,\(^8–12\) photoelectrochemical cell,\(^13–16\) capacitive device,\(^17–21\) and photovoltaic\(^22–26\) applications. According to the Shockley–Queisser limit, \(\sim30\%\) power conversion efficiency (PCE) could be obtained from the single-junction CuO-based solar cells.\(^20\)

A suitable n-type material is required with the CuO absorber layer for the formation of a single p–n junction CuO-based solar cell. ZnO is an extensively used electron transport material in low-cost highly efficient solar cells.\(^27–29\) It is an earth-abundant, non-toxic, and wide band gap n-type IMO with high electron mobility.\(^30\) The energy band alignment of n-type ZnO is suitably matched with p-type CuO.\(^31,32\) Therefore, ZnO could be a promising electron transport layer (ETL) for the CuO-based solar cells. Furthermore, few research groups have experimented on ZnO-CuO-based solar cells in recent years, but the reported efficiency of these solar cells is very low.\(^33–36\) There are many effective ways to improve the performance of the heterojunction solar cell (HSC) like band gap tuning\(^37,38\) band alignment engineering,\(^39\) crystallinity improvement of the absorber layer,\(^40\) use of charge-selective layers,\(^41–43\) etc. Kapile and co-workers have worked on the band alignment engineering of the ZnO-CuO-based hetero-
junction solar cell and observed a considerable performance improvement. Kuddus et al. have researched on the performance enhancement of the ZnO-CuO-based heterojunction solar cell through the band gap tuning of CuO using a silicon nanoparticle (Si-NP) dopant. Naveena et al. have also experimented on the band gap tuning of CuO using an ytterbium (Yb) dopant and observed a significant role of this technique on the performance enhancement of the ZnO-CuO-based heterojunction solar cell.

The introduction of a hole transport layer (HTL) has been found to be an effective technique for the performance enhancement of the heterojunction solar cell. Recently, attractive featured transition metal oxides (TMOs) such as nickel oxide (NiO), tungsten trioxide (WO3), cuprous oxide (Cu2O), molybdenum trioxide (MoO3), and vanadium pentoxide (V2O5) have been used in the different organic and inorganic solar cells as the HTL. The low cost, contact with the active layer, high optical transparency in the visible range, good stability at ambient conditions, a wide range of band alignments, and feasible deposition by a facile solution process are the most fascinating features of these TMOs. Kaphle et al. have found a significant performance enhancement after the introduction of the MoO3 HTL in a practical structure of the ZnO-CuO-based solar cell. Lam has conducted a numerical study on the planar ZnO-CuO based-solar cell using the Cu2O HTL and reported a maximum achievable PCE of 12.18%. The valence band maximum (VBM) and the conduction band maximum (CBM) of V2O5 are suitably matched with the VBM and the CBM of CuO, which could accelerate the hole transport from the CuO absorber layer to the output terminal of the solar cell as well as block the photogenerated electrons at the CuO/V2O5 interface. Therefore, V2O5 could be a potential hole extracting layer for the ZnO/CuO-based solar cell.

Simulation is a widely accepted technique for device modeling, performance analysis, and understanding the overall device physics of the designed heterojunction solar cells. The key advantage of the numerical simulation is that the effect of different physical parameters of the materials on the device performance can be easily investigated. Such a type of investigation provides the guidelines to obtain optimum performance from a designed solar cell without fabricating it in the laboratory. For numerical simulation of heterojunction solar cells, a solar cell capacitance simulator (SCAPS), a solar cell capacitance simulator (SCAPS), analysis of microelectronic and photonic structures (AMPS), and Silvaco TCAD have been popularly used in recent years.

In this research, low-cost CuO-ZnO-based solar cells have been successfully fabricated using the spin coating technique. The solution-processed V2O5 HTL has been introduced in the CuO-ZnO-based solar cells. V2O5 has dramatically boosted the Voc and device performance. A numerical simulation on the defect features of the CuO absorber layer has also been conducted using the SCAPS simulation program. A maximum PCE of 1.69% has been recorded from the experimentally fabricated CuO-ZnO-based solar cell.

2. RESULTS AND DISCUSSION

2.1. Structural Properties. ZnO, CuO, and V2O5 thin films were fabricated using prepared ZnO, CuO, and V2O5 solutions (Section 4.2), respectively, and annealed at a temperature of 300 °C for 1 h in open air. XRD analysis of these films was done for the confirmation of ZnO, CuO, and V2O5, shown in Figure 1. From Figure 1, it is seen that the peaks at 2θ values of 31.84, 34.32, 36.38, 47.64, and 56.7° correspond to (100), (002), (101), (102), and (110) planes evident to the ZnO wurtzite crystal structure (JCPDS card no. 36-1451). CuO confirmation peaks at 32.5, 35.4, 38.7, and 48.7° correspond to (110), (002), (111), and (202) planes, and these planes have revealed the monoclinic crystal structure of CuO (JCPDS card no. 48-1548). In Figure 1, the sharp peak at a 2θ value of 20.12° corresponds to the (010) plane, which indicates the orthorhombic structure of V2O5 (JCPDS card no. 01-076-1803). The fabricated V2O5 film was also polycrystalline in nature. There are some peaks of V2O5 that are not clear in Figure 1 due to their low intensity.

The cell performance is strongly dependent on the surface morphology of the HTL that defines the interface between the absorber layer (CuO) and the back metal contact (Ag). SEM was performed to analyze the top surface morphology of two-layer V2O5 on top of the CuO-ZnO-based HSC, shown in Figure 2. A compressed, quite uniform, and almost pinhole-free surface of V2O5 on CuO is observed from Figure 2. A similar surface is expected to form at the back side of V2O5 at the surface of V2O5 on top of the CuO-ZnO-based HSC, shown in Figure 2. A compressed, quite uniform, and almost pinhole-free surface of V2O5 on CuO is observed from Figure 2. A similar surface is expected to form at the back side of V2O5 at the
CuO/V2O5 interface. Therefore, the CuO/V2O5 interface as well as the V2O5/Ag interface could help to form a good ohmic contact between the CuO absorber layer and the Ag back contact, which in turn will reduce the device series resistance.

The chemical compositions present in the CuO-ZnO-based HSC with the two-layer V2O5 HTL were analyzed using EDS, which are listed in Table 1. Table 1 indicates that oxygen (O), vanadium (V), copper (Cu), zinc (Zn), indium (In), and tin (Sn) were present in the CuO-ZnO-based HSC with the two-layer V2O5 HTL.

Table 1. Elemental Composition of the CuO-ZnO-Based HSC with the Two-Layer V2O5 HTL Observed from the EDS Spectra

| element | weight % | atomic % |
|---------|----------|----------|
| Zn      | 12.07    | 8.17     |
| Cu      | 45.73    | 31.85    |
| V       | 10.51    | 9.13     |
| O       | 16.28    | 45.08    |
| In      | 1.29     | 0.5      |
| Sn      | 14.13    | 5.27     |

2.2. Optical Properties. The optical transmittance spectra of V2O5 thin films annealed at 250, 300, and 350 °C in open air for 1 h are illustrated in Figure 3. It is observed that the transmittance increases between the wavelength of 520 and 1100 nm with increasing the annealing temperature, which is due to the thermochromism property of V2O5.68 The optical band gap has been found to be around 2.45, 2.4, and 2.3 eV from the V2O5 thin films annealed at 250, 300, and 350 °C, respectively, by using the Tauc formula, \((\alpha h\nu)^2 = \epsilon(h\nu - E_g)\), which is depicted in Figure 3b. The variation of the band gap might be due to the change of stoichiometry of V2O5 with the temperature change.

2.3. Electrical Properties. The energy band diagram and the schematic diagram of the CuO-ZnO-based HSCs with the V2O5 HTL are depicted in Figure 4. The junction property of the CuO-ZnO-based HSC with two-layer V2O5 was characterized by capacitance−voltage (C−V) measurement using an impedance analyzer. Figure 5 delineates the Mott−Schottky plot of the CuO-ZnO-based HSC with the two-layer V2O5 HTL. The ascertainment of the built-in potential (\(\psi_{bi}\)) of the CuO-ZnO-based HSC with V2O5 was done by fitting and extrapolating the linear portion of the plots, and \(\psi_{bi}\) was found to vary from 0.68 to 0.76 V at frequencies of 500 Hz, 1 kHz, and 10 kHz. According to the p−n junction solar cell theory, the built-in potential is nearly equal to the open-circuit voltage (\(V_{oc}\)) under a sunlight illumination of 100 mW/cm².69 The J−V characteristics of the fabricated CuO-ZnO-based HSCs with and without V2O5 were investigated under an illumination of 1.5 AM (100 mW/cm²) simulated sunlight conditions. As the main focus of this research is to observe the...
influence of the solution-processed V$_2$O$_5$ HTL on the performance of CuO-ZnO-based HSCs, different parameters of V$_2$O$_5$ like thickness and deposition temperature should be optimized. Here, the thickness of V$_2$O$_5$ was experimented with the number of deposited layers keeping deposition temperature constant. Figure 6 depicts the $J$–$V$ characteristics of CuO-ZnO-based HSCs without the V$_2$O$_5$ HTL and with one-layer, two-layer, and three-layer V$_2$O$_5$ HTLs.

Table 2. The Comparison of Photovoltaic Parameters of CuO-ZnO-Based Inorganic Thin-Film HSCs with Different Thicknesses of the V$_2$O$_5$ Hole Transport Layer

| V$_2$O$_5$ layer number | $V_{OC}$ (V) | $J_{SC}$ (mA/cm$^2$) | FF (%) | PCE (%) | $R_s$ (Ω cm$^2$) |
|------------------------|--------------|----------------------|--------|---------|-----------------|
| 0                      | 0.389        | 2.54                 | 34     | 0.332   | 122             |
| 1                      | 0.475        | 5.18                 | 42     | 1.029   | 44              |
| 2                      | 0.62         | 6.78                 | 40.2   | 1.69    | 28              |
| 3                      | 0.515        | 5.87                 | 35     | 1.054   | 57              |

Generally, the series resistance ($R_s$) of the solar cell comes from the absorber layer and contact electrodes.$^{71}$ The insertion of the V$_2$O$_5$ HTL creates two interfaces CuO/V$_2$O$_5$ and V$_2$O$_5$/Ag, and these interfaces might contribute to the lowering of the series resistance of the TTO/ZnO/CuO/V$_2$O$_5$/Ag heterostructure. The series resistance ($R_s$) of the CuO-ZnO-based HSCs without and with the V$_2$O$_5$ HTL was estimated from the slope of the $J$–$V$ curve at a current density ($J$) of 0 mA/cm$^2$ and is listed in Table 2. The estimated series resistance indicates that the two-layer V$_2$O$_5$ HTL remarkably reduced the contact resistance around three times.

2.4. Theoretical Analysis of Defect Properties of the CuO Absorber Layer. The non-radiative Shockley–Read–Hall (SRH) recombination is the main factor that is responsible for most of the power loss in photovoltaics.$^{72}$ The presence of the deep level defect in the absorber layer is the primary source of the SRH recombination.$^{73}$ High defect density in the absorber layer reduces the photogenerated carrier lifetime and device performance. SRH recombination can be defined by the following equation

$$R_{SRH} = \frac{np - n_i^2}{\tau (p + n - 2n_i \cosh (\frac{k_i - k_f}{kT}))}$$  \hspace{1cm} (1)

where $\tau$, $E_i$, $k$, and $T$ are the carrier lifetime, defect activation energy level, Boltzmann constant, and device working temperature, respectively.

Carrier lifetime can be estimated using the following equation

$$\tau = \frac{1}{\sigma N_v \times V_{th}}$$  \hspace{1cm} (2)

where $\sigma$, $N_v$, and $V_{th}$ are the carrier capture cross section, defect density, and thermal velocity of the carrier, respectively.

The fabricated CuO-ZnO-based HSC with two-layer V$_2$O$_5$ was theoretically validated using the SCAPS simulation tool. The $J$–$V$ characteristics and quantum efficiency curves of the theoretically validated device are depicted in Figure S2. The whole simulation study was conducted using the default parameters listed in Table S1. The CuO absorber layer defect properties were numerically analyzed using the SCAPS simulation program. The photovoltaic performance of the theoretically validated CuO-ZnO-based HSC was studied by varying the CuO absorber layer thickness from 400 to 1400 nm and CuO deep level defect density from $10^{10}$ to $10^{18}$ cm$^{-3}$, while the hole capture cross section ($\sigma_p$) and defect activation energy ($E_A$) were kept constant at $10^{-35}$ cm$^2$ and 0.6 eV, respectively. It is observed from Figure 7c that the $V_{OC}$ decreases with the increase in defect density above $10^{15}$ cm$^{-3}$. Figure 7a indicates that the SRH recombination increases with the increase in defect density, which in turn increases the non-radiative loss of the V$_2$O$_5$. It is also obvious from Figure 7b that the carrier lifetime at the CuO absorber layer decreases with the defect density. Figure 7 indicates that the defect density up to $10^{11}$ cm$^{-3}$ is tolerable and above which PCE is drastically reduced with the defect density.

From eqs 1 and 2, it is clear that the $E_A$ and hole capture cross section $\sigma_p$ of the CuO absorber layer have a considerable effect on the SRH recombination and the carrier lifetime. It is observed from Figure 8a that the SRH recombination increases with the $E_A$. As shown in Figure 8b, the carrier lifetime decreases with $\sigma_p$. The performance of the theoretically...
validated CuO-ZnO-based HSC was investigated by varying $E_A$ from 0.1 to 0.6 eV (above $E_V$) and $\sigma_P$ from $10^{-17}$ to $10^{-8}$ cm$^2$, while the defect density of the CuO layer was kept constant at $10^{16}$ cm$^{-3}$. As shown in Figure 8c,f, the $V_{OC}$ and PCE are almost independent of the $\sigma_P$ till an $E_A$ of 0.2 eV, while the $V_{OC}$ and PCE decrease with increasing $E_A$ above 0.2 eV. The $E_A$-assisted decrement of the $V_{OC}$ and PCE is consistent with the variation of SRH recombination, which is shown in Figure 8a.

The effect of $\sigma_P$ was found to be dominant at $E_A > 0.45$ eV. Figure S3 indicates that the optical loss at the front surface of the CuO-ZnO-based HSC is another factor that is responsible for the low performance besides the defect properties of the CuO absorber layer.
3. CONCLUSIONS

Low-cost environment-friendly CuO-ZnO-based solar cells have been successfully fabricated using a facile chemical route. The solution-processed V$_2$O$_5$ HTL has been inserted in the heterostructure of the CuO-ZnO solar cells using the spin coating technique. The $V_{oc}$ was enhanced up to 231 mV after deposition of the V$_2$O$_5$, HTL in between the CuO and Ag electrode, which indicates that the V$_2$O$_5$ HTL has significantly reduced the non-radiative SRH recombination at the CuO surface. The performance of the CuO-ZnO-based solar cells has also been numerically investigated using the SCAPS simulation tool. It is obvious from the numerical investigations that the CuO absorber layer defect parameters like $N_d$, $E_A$, and $\sigma_p$ have a huge impact on the device performance. The overall findings indicate that solution-processed V$_2$O$_5$ could be a potential HTL for the low-cost CuO-ZnO-based solar cells.

4. EXPERIMENTAL SECTION

4.1. Materials. Diethanolamine (DEA, ≥98.0%), ethanol (CH$_3$CH$_2$OH, 96%), zinc acetate dihydrate [Zn(CH$_3$COO)$_2$·2H$_2$O, 99.999%], copper acetate monohydrate [Cu(CH$_3$COO)$_2$·H$_2$O, ≥99%], vanadium pentoxide (V$_2$O$_5$, 99.95%), and indium tin oxide (ITO)-coated glass substrates (surface resistivity of around 8–12 $\Omega$/sq) were purchased from Sigma-Aldrich. All chemicals were used without further purification.

4.2. Solution Preparation. The ZnO solution was prepared by dissolving 0.65 g of zinc acetate dihydrate in 20 mL of ethanol. Then, the solution was stirred with the help of a magnetic stirrer at 40 °C for the next 10 min. After that, 1 mL of diethanolamine (DEA) as a stabilizer was added to this solution, and the solution was also stirred for the next 30 min at a temperature of 40 °C. Hence, a homogeneous solution of ZnO was obtained. Applying a similar process, 0.6 g of copper acetate monohydrate, 2 mL of DEA, and 20 mL of ethanol were used to prepare the CuO solution. By a similar process, the V$_2$O$_5$ solution was also prepared with 0.25 g of vanadium oxide powder, 2 mL of DEA, and 20 mL of ethanol.

4.3. Cell Fabrication. The ITO glass substrates were used to fabricate the HSCs. Before fabrication, the ITO glass substrates were cleaned in an ultrasonic vibrator using acetone, isopropanol, and distilled water for 15 min in sequence. Then, the cleaned ITO glass substrate was placed on a spin coating system, and 10 µL of the ZnO solution was drop cast on the ITO glass substrate and spun for 30s at 1500 rpm. The film was preheated at 150 °C for 10 min, and this process was repeated for the deposition of each layer of ZnO. A similar process was adopted for the deposition of the absorber layer of CuO on the ZnO layer. Finally, after annealing at 300 °C for 1 h in open air, the ZnO-CuO-based HSC without an HTL was fabricated. A patterned Ag paste was used to make the back-contact of the solar cells. Through a similar process, the ZnO-CuO-based heterojunction solar cell with an HTL was fabricated. The prepared solution of V$_2$O$_5$ was deposited on the CuO layer as an HTL by spin coating, and finally, the film was annealed at 300 °C for 1 h in open air. The fabrication flow diagram is shown in Figure 9.

4.4. Characterization. The crystallographic analysis of XRD patterns of ZnO, CuO, and V$_2$O$_5$ thin films was done using an X-ray diffractometer (GBC, eMMMA) with monochromatic Cu K$_\alpha$ radiation having a wavelength of 1.540598 Å. SEM images were also taken using a scanning electron microscope (SEM, Zeiss, EVO 18). Energy-dispersive X-ray spectroscopy (EDAX, AMETEK) in SEM was performed to analyze chemical compositions present in ITO/ZnO/CuO/V$_2$O$_5$. Transmittance spectra of V$_2$O$_5$ thin films were measured by T-60 UV-visible spectroscopy, and current density versus voltage ($J$–$V$) of the fabricated solar cells was measured using a Keithley 2400 source meter under an illumination of 1.5 AM simulated light. All characterizations were conducted at room temperature (∼27 °C) and a relative humidity (RH) of ∼50%.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c00678.

Physical parameters used in the simulation (Table S1); absorption coefficients of ZnO, CuO, and V$_2$O$_5$ (Figure S1); $J$–$V$ characteristics and QE curve of the theoretically validated CuO-ZnO-based HSC (Figure S1); and a flow diagram of the CuO-ZnO-based heterojunction solar cell fabrication process (Figure 9).
S2); and J–V characteristics of the theoretically validated CuO-ZnO-based HSC for various optical losses (Figure S3) (PDF)

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Notes
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