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Photoelectrochemical solar cells enactment of ternary $MoS_2/WS_2/Cu_2O$ based photocathode under visible-light irradiation

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Abstract

The development of inexpensive and effective working and counter electrodes is of crucial importance for strongly enhancing the performance in photoelectrochemical solar cell. Here we first off report a rational and cost-effective design of a ternary photocathode that embeds Cu_2O between two dimensional transition metal dichalcogenides by chemical vapor deposition to achieve improved PEC performance. Ternary hetero-epitaxial growth was attainable by growing of MoS_2/WS_2 on the Cu_2O hybrid by a partial thermal oxidation process. $MoS_2/WS_2/Cu_2O$ is indeed conducive to boost the solar light absorption, excite the hot electron-hole pairs and elevate ultrafast spatial transfer and separation of photogenerated holes and electrons, resulting in remarkable photocurrent 0.78mA, a sixteen-fold momentous improvement in maximum photoconversion efficiency from 0.036% to 0.58% enhancement than that of pristine Cu_2O. Therefore, this work may stimulate continued interest in the design of high-efficiency, low-cost hybrid photoelectrodes that can replace stately Cu_2O in photoelectrochemical solar cell applications. The materials were structurally characterized using X-ray diffraction, Ftir and photoelectrochemical-related dimensions under visible-light irradiation. Under illumination the PEC performance of the ternary hybrid electrode was investigated using current-voltage test. The mechanisms responsible for the enhanced PEC behavior of the $MoS_2/WS_2/Cu_2O$ ternary hybrid are discussed in detail.

Keywords: annealing, etching, thermal oxidation; cuprous oxide; hetero-structure; I-V characteristic, photoelectrochemical

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1. Introduction

The utilization of inexhaustible and clean energy such as hydrogen energy is an important issue for coping with the global energy crisis and environmental problems [1]. Photoelectrochemical (PEC) water splitting is a promising approach for using and converting solar energy to produce hydrogen fuel.In the ongoing effort to develop effective photoelectrodes, the greatest challenge currently facing researchers is to find highly efficient, inexpensive and terrestrially abundant photoelectrodes [2]. Among various metal oxides, cuprous oxide (Cu_2O) as photocathode which exhibits p-type conductivity dominated by copper vacancies has drawn intensively attention in the application of photoelectrochemical water splitting [3]. Its advantages include a suitable direct bandgap of 2.0 eV for PEC, non-toxicity and, more importantly, favorable conduction and valence bands lie negative and positive for H_2 and O_2 evolution potentials, respectively [4,5]. However, the low charge transfer kinetics in Cu_2O and poor stability in aqueous solution greatly hinder the utilization efficiency in water splitting [6]. To date, many efforts have been made to eliminate the above problems, including: Coupling by conformal coating and decoration by noble metal semiconductors[6,7,8]. Unfortunately, for single improvement projects Cu_2O is not as efficient as expected. The high cost of efficient photocatalysts is also an unavoidable problem. Therefore, the development of multi-component systems integrating inexpensive high efficiency light absorbers and high turnover catalysts is a promising approach [9,10]

To improve the PEC efficiency of photoelectrodes, fabrication of semiconductors with heterostructures and compound or elemental doping have been extensively investigated [11-12], due to the different interactions between different semiconductor materials. Among them (Cu_2O) has often been considered as the newest candidate for photocathode [13,14]. Cu_2O is a typical p-type semiconductor with a bandgap of about 2 eV in the AM 1.5 spectrum, the theoretical photocurrent for water splitting of -14.7 $mAcm^{-2}$ and the solar to hydrogen conversion efficiency of 18.1 % [14].

In addition, it is scalable, abundant on earth, environmentally friendly and compatible with inexpensive synthetic processes. This is an essential prerequisite for covering the world's energy needs in the terawatt range [15]. Despite the above advantages, the practical application of Cu_2O in PEC processes is still limited by two main drawbacks: (1) The skyscraping recombination rate of photogenerated electron-hole carriers is due in part to the mismatch between the electron diffusion length (typically 20–100 nm) and the optical absorption depth is [16]; (2) Loss of photostability due to self-photocorrosion in the electrolyte [17]. Copper (I) oxide (Cu_2O) pattern engineering has been reported to effectively address the above limitations. Currently, most Cu_2O-based photocathodes for PEC water splitting are usually built on Cu_2O films due to the simple fabrication process with low photoelectric conversion efficiency [18,19]. In contrast, its Cu_2O nanowire/nanorod-based counterparts show significantly improved

efficiency. This is mainly attributed to the transport of photogenerated charge carriers, a larger surface area for more efficient light collection and fast charge transfer at the interface, more efficient separation and electrochemical reactions [20-21]. Besides structural engineering, heterojunction engineering is widely recognized as another effective strategy to improve the PEC water splitting performance of Cu_2O by efficiently separating photogenerated charge carriers [22-23]. The Cu_2O-bonded semiconductor is not only a key element in the formation of pn junctions, but in some cases also acts as a protective layer that retards sample corrosion [24-25]. Despite tremendous efforts, Cu_2O-based photocathodes remain a challenge, and combining this two strategies described above could lead to a novel and efficient a Cu_2O-based photocathode can be developed with potential applications in PEC water splitting. This suggests that the ongoing need to study the photocathode further may go further.

This work deals with PEC investigations of Cu_2O layers produced by partial thermal oxidation. Copper forms two different oxides. Copper forms two different oxides, Copper II oxide (CuO) with a bandgap of 1.21–1.51 eV [28,29] absorbs the entire visible range, whereas Copper I oxide (Cu_2O) with a direct bandgap of 2.1 eV [26,27], it strongly absorbs only at wavelengths below 600 nm. Other reasons for choosing Cu_2O as the material in this study are (a) its low production cost, (b) its natural occurrence, (c) non-toxicity, (d) stability, and (e) fairly good electrical characteristics. The effects of hybrid ternary deposition of $MoS_2/WS_2/Cu_2O$ film's PEC behavior were studied. In addition, the films were also characterized with respect to phase and structure analysis using XRD, I-V characteristic curve and FTIR, respectively.

2. Materials and methods

2.1 Synthesis of Cu_2O thin film

Commercial pure copper (99.98%) in foil form (0.1 mm thick) was sliced into criterion size 2 cm x 2 cm wafers. The sample was smoothened by pickling the edges of the bottle, dipped in dilute nitric acid, rinsed well with distilled water several times, and then dried to remove impurities on the membrane surface. After cleaning, the copper film was thermally oxidized by furnace annealing in air. Starting from room temperature (RT) to 450°C the oxidation temperature was regimented over a wide range. The heating rate was about 10°C./min, and once the preferred maximum temperature was reached, it was held for 30 minutes to allow copper oxide to form. After oxide formation, the furnace was allowed to cool for 2 hours. Slow cooling was maintained to minimize potential thermal stress and film cracking.

2.2 Synthesis of $MoS_2/WS_2/Cu_2O$ hybrid film

The method used to grow MoS_2 was chemical vapor deposition (CVD) [30, 31]. The substrate was subjected to specific temperature and pressure conditions to chemically react one or more precursors on

the surface of the substrate to produce high-quality, large-area thin films.

The application of CVD in monolayer TMD fabrication begins with the growth of WS_2. The heating rate was set at 10°C per minute, the growth temperature was 650°C and the temperature was held for 30 minutes. When the temperature dropped to 400°C, the lid was opened

A WS_2 structure was obtained when the temperature was lowered to room temperature. The experimental process is shown in Figure 1.0. CVD can efficiently produce monolayer and multilayerMoS_2/WS_2. We were able to grow thin films and produce high-quality single-crystal materials uniformly distributed over a large area. This was useful in the production of later optoelectronic components.

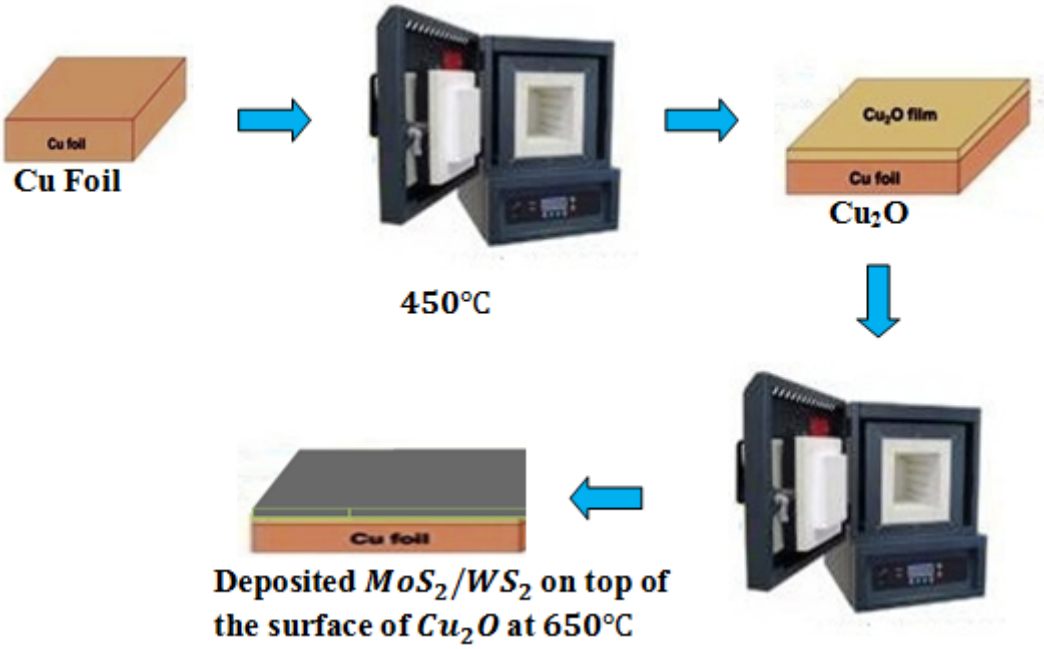

Figure 1. Schematic of a growing $\boldsymbol{MoS_2/WS_2/Cu_2O}$ film.

Photoelectrochemical tests

For this purpose, $MoS_2/WS_2/Cu_2O$and Cu electrode were placed and immersed in a transparent plastic container. To prepare the electrolyte, the 1g of NaCl powders was mixed with 25ml of distilled water and stirred gently until the electrolyte was completely dissolved. The PEC performance of the hybrid electrodes was evaluated using Current-Voltage measurements (Figure 2). Photoelectrochemical studies were performed using a two-electrode electrochemical system. A working electrode ($MoS_2/WS_2/Cu_2O$) and a copper plate were used as counter electrodes, respectively. A multimeter was used to monitor the electrical pathways of the electrode photocurrent and photovoltage under illumination (AM 1.5 G) in a potential window using 1 g of NaCl electrolyte as a mediator between the two electrodes. The assembly system described in this work provides a simple and novel method for synthesizing thin-film materials ready for practical applications such as hydrogen production and photoelectrochemical solar cells.

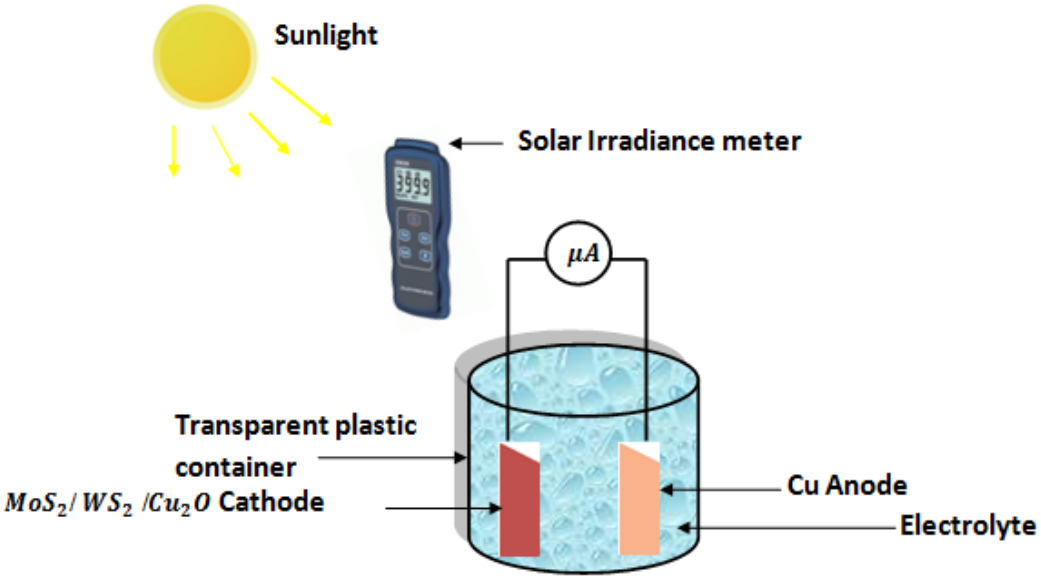

Figure 2: Illustration of the fabricated Cu –$MoS_2/WS_2/Cu_2O$ Photoelectrochemical solar cell.

Results and discussion

Structural analysis

The purity, phase composition, crystallinity and lattice parameters of the synthesized materials were estimated by x-ray diffraction (XRD) analysis. The X-ray data were collected in the 2θ range of 10 - 70° at a sampling rate of 0.02° per second. Figure 3 shows that the XRD pattern of heteroepitaxial growth of$MoS_2/WS_2/Cu_2O$ thin film deposited at different thermally oxidation temperatures. XRD patterns indicate that the deposited Cu_2O films are polycrystalline in nature and belong to cubic structure.

In addition, all characteristic smooth tilt peaks from the ternary sample were fully inspected and where the diffraction peaks are at 2θ values of 29.01°, 36.52°, 42.11° and 61.11° corresponding to (110), (111), (200) and (220) diffraction plane of Cu_2O (JCPDS card no. 05-0667), respectively, were detected [32,33,34,65]. The observed peaks at 36.1° and 55.2° for thermally oxidized WS_2 correspond to (102), (103) [35, 36]. The peaks observed at 46.1°, 61.7° for thermally oxidized MoS_2 correspond (104) (008) [37]. Presence of a reflection at 39.3° attributed to platinum(111) [40,41]. The 33.3° peak corresponds to CuO(110) [42]. MoO_2 also shows peaks that correspond to (110) at 24.7°, (104) [43,44]. Copper Cu (111) and (200) were observed at 43.8°, 51.2° for Cu, in addition to the peaks corresponding to the Cu_2O [38,39]. Table 1 displayed the qualitative analysis result of the ternary sample.

FT-IR spectra analysis

FT-IR analysis (Figure 4) was performed to analyze the structural and chemical properties of bare MoS_2, WS_2, and Cu_2O thin films. On the other hand1477 cm^{-1}, and 1659 cm^{-1}, are the broad absorption bands attributed to MoS_2. Also, the bands at 914 cm^{-1} are due to S-S bond [45].

Qualitative Analysis Results

S.No.	Phase name	Formula	Figure of merit
1	Cuprite	Cu_2O	0.930
2	Molybdenite-2H	MoS_2	2.900
3	Platinum	Pt	1.029
4	Quartz	SiO_2	0.820

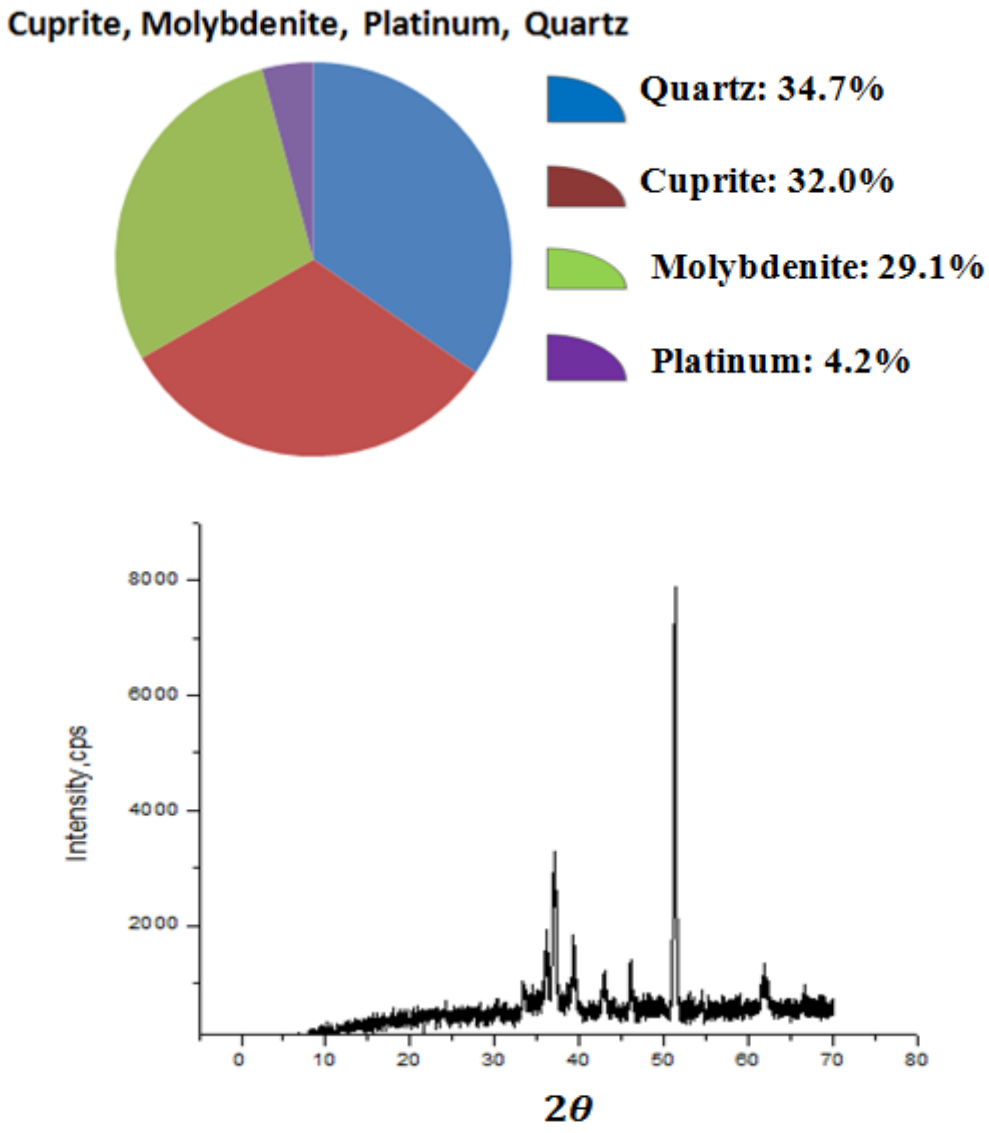

Figure 3. XRD pattern of$MoS_2/WS_2/Cu_2O$

FTIR spectrum of MoS_2 revealed a broad band at 1640cm^{1} which are attributed to bending and stretching vibration of H–O–H [45]. Furthermore, stronger bands are observed at 1741 and 1730 cm^{1}due to the C=O stretches can be observed [46]. A broad shoulder band ranging from 1360 and 890 cm^{1} correspond to the stretching vibration of the terminal NH groups at the aromatic ring defects [47, 48]. The presence of oxygen-related bonds was due to the presence of sharp bonds ~3620 cm^{-1} [47, 48]. A strong peak at

685 cm^{-1}indicates the presence of W-S related compound. The peak at 2950 and 3450 cm^{-1}1 are assigned to the -OH groups [47, 48], and 3450, 3150 cm^{-1} are broad absorption bands assigned to WS_2 [49, 50]. The appearance of peak at between 876 and 825 cm^{-1}belong to Cu(II)-O. The peak at 1658 cm^{-1} is due to the hydroxyl related compound [44, 45]. 2850 $cm^{-1}$is assigned to symmetric C-H stretch bond vibration while 1470cm^{-1}attributed to the presence of CH_2, CH_3 at 2850cm^{-1}. From the illustration in Figure 4 the characteristic peak of Cu_2O ((630cm^1) is observed shift (towards lower wavenumber) after the incorporation of MoS_2 and WS_2, which indicated that there was an interaction between MoS_2, Cu_2O and WS_2. From the spectra, all the characteristic peaks of MoS_2, Cu_2O and WS_2 appeared in the $MoS_2/WS_2/Cu_2O$, which is consistent with the XPS spectra.

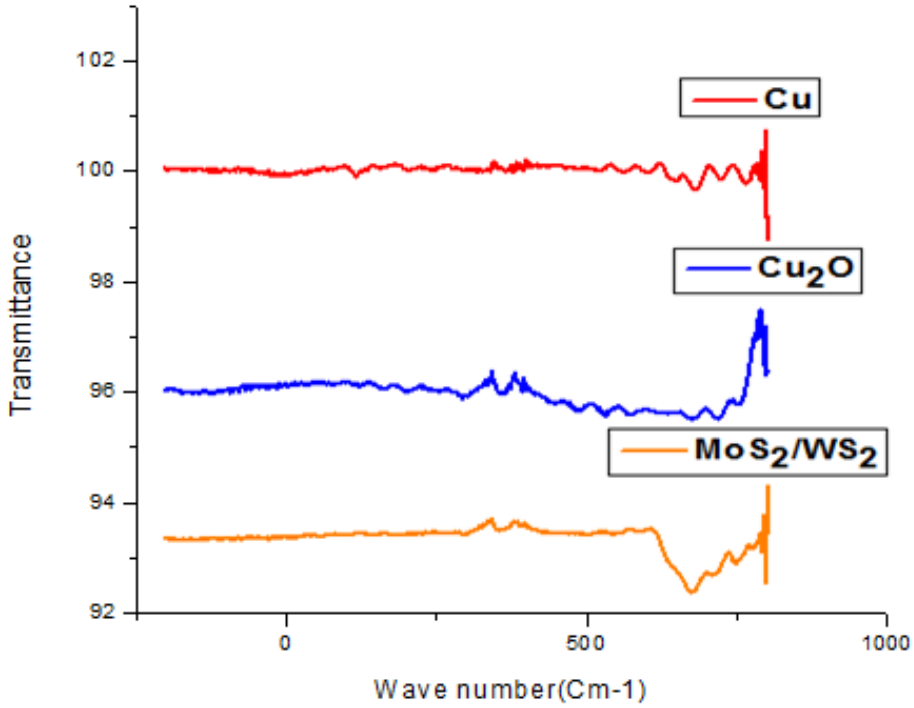

Figure 4. FTIR spectra of $MoS_2/WS_2/Cu_2O$

I-V Curve analysis

The performances of the Cu_2O films were measured in terms of photocurrent through I-V Curve analysis the efficiency, maximum power, photo voltage and photocurrent were obtained underneath illumination as outline in Table 2.0. The synthesized Cu-Cu_2O and Cu- $MoS_2/WS_2/Cu_2O$ PEC solar cell exhibited characteristic curves of many extrinsic parameters followed by transition power efficiency derived from figure 4 and figure 5. For the prototype analysis, the calculated extrinsic parameters of the prepared samples Cu-Cu_2O and $MoS_2/WS_2/Cu_2O$are given in table 2.0. To study the solar cell parameters, examine two different graphs for two different samples to test the photoresponse and photovoltage of the electrodes under illumination. In table 2.0 for the synthesized $MoS_2/WS_2/Cu_2O$, we find that the

deposition of 2D materials enhances the photoresponse while increasing the sample efficiency. It also acts as an absorption layer to generate charge carriers (electrons and holes) under solar radiation. Some results of Isc, Voc, and η compared with other studies in the literature, as shown in Table 3 below. Table 2.0: The photocurrent, maximum power, efficiency and photo voltage of different measurements of Cu-Cu_2O and Cu- $MoS_2/WS_2/Cu_2O$ photoelectrochemical solar cell

S/N	Sample	$I_{sc}(mA)$	$V_{oc}(mV)$	$\eta(\%)$
1	Cu_2O	0.14	11.0	0.036
2	MoS_2/WS_2	0.78	50	0.58

The first row in table 2.0 is forCu-Cu_2O synthesized using thermal oxidation method while the subsequent row is for $MoS_2/WS_2/g/Cu_2O$ layer deposited by partially thermal oxidation method.

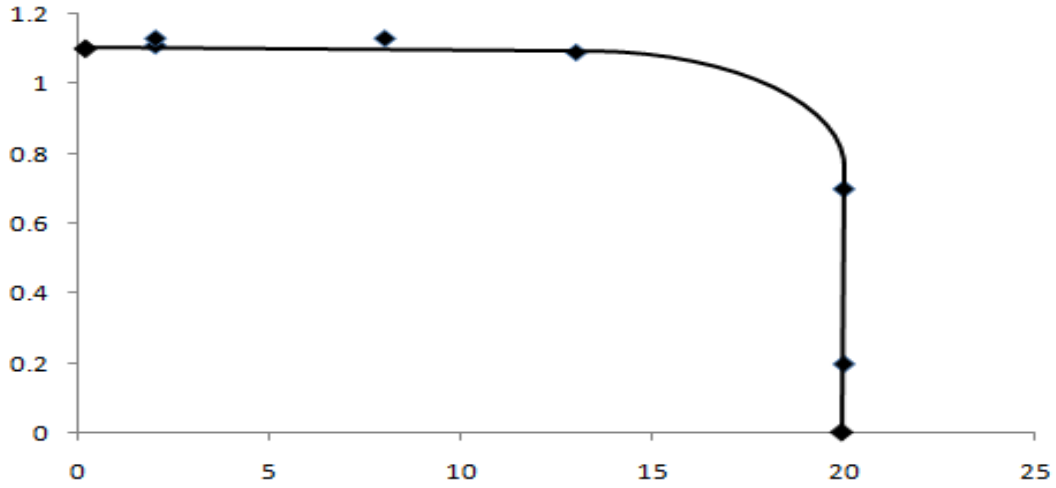

Figure. 4: The graph of Cu-Cu_2O photoelectrochemical solar cell before surface modification

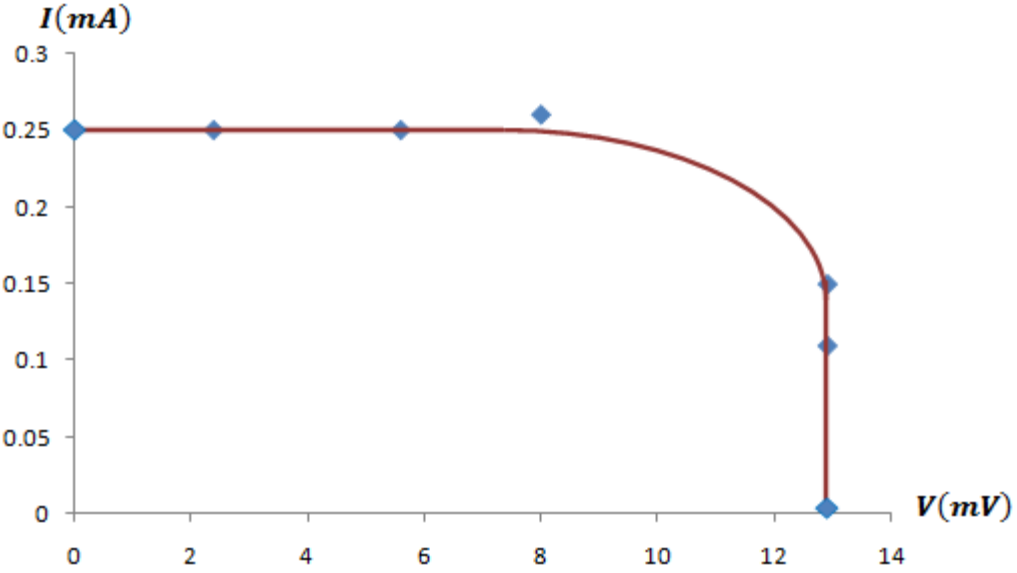

Figure. 5: The graph of Cu- $MoS_2/WS_2/Cu_2O$ photoelectrochemical solar cell after surface modification

Table 3: Comparison of Findings with other Works in Literature

Authors(Year)	Structures	Method	Efficiency	findings	References
Wilman et al (2011)	Cu_2O/Al-doped	Electro-deposition	O.60%		[51]
R. P. Wijesundera *(2010)*	Ti/CuO/Cu_2O/Au	Electro-deposition	0.02%	*FF, Isc,Voc*	[52]
Katayama et al (2004)	Cu_2O/ZnO/ITO	Electro-deposition	0.117%	*FF, Isc,Voc*	[53]
Septina et al (2011)	Cu_2O/AZO	Electro-deposition	0.60%	*FF, Isc,Voc*	[54]
Seyed, A.J (2013)	Cu_2O	Electro-deposition	0.082%	*FF, Isc,Voc*	[55]
Y.-K. Hsu, et al *(2015)*	Cu_2O	Electro-deposition	0.42	*FF, Isc,Voc*	[56]
Abdu Y*, (2017)*	Cu_2O	Thermal oxidation	0.08	*FF, Isc,Voc*	[57]
Vijayaraghavan, et al *(2018)*	CdTe	SPD technique	0.062	*FF, Isc,Voc*	[58]
Roza, et al *(2014)*	ZnO	Hydrothermal	0.050	*FF, Isc,Voc*	[59]
Abdurrahman, M *,(2019)*	Cu_2O	Thermal oxidation	0.046	*FF, Isc,Voc*	[60]
Sutripto, et al *(2019)*	CdO	chemical method	0.21	*FF, Isc,Voc*	[61]
TadatsuguMinami, *et al(2016)*	Zn_2GeO_4	Thin film deposition	0.12	*FF, Isc,Voc*	[62]
TadatsuguMinami, *et al(2016)*	Zn_2SiO_4	Thin film deposition	0.03	*FF, Isc,Voc*	[62]
Tadatsugu Minami, *et al(2016)*	$ZnSnO_3$	Thin film deposition	0.01	*FF, Isc,Voc*	[62]
This work		Thermal oxidation	0.58	*FF, Isc,Voc*	
Abdurrahman, M*,(2022)*	Cu_2O	Thermal oxidation	4.80	*FF, Isc,Voc*	[63]
Yin X et al*,(2018)*	FeOOH/TiO_2/ $BiVO_4$-Cu_2O	Electrochemical deposition	0.46%	*FF, Isc,Voc*	[64]

Conclusion

In this work, ternary hybrid composed ofWS_2, $MoS_2$and Cu_2O thin films are used to demonstrate a simple and inexpensive method to improve the conversion efficiency of photoelectrochemical solar cell applications. The MoS_2/WS_2binary hybrid was prepared by partial thermal oxidation processing of WS_2 andMoS_2in a high temperature closed furnace, optical and morphology properties of the sample was also investigated. The epitaxial growth of WS_2 and that of MoS_2on the surfaces of Cu_2O thin film occurred during the partial thermal oxidation process. The Full structure and PEC analysis indicate that the increased PEC activity could be attributed to the synergistic effect between the two 2D materials. In addition, 2D materials exhibit a passivation effect that not only improves photo-excitation voltage and current by slowing the recombination rate of charge carriers, but also increases surface photoelectrochemical to improve the photocurrent by separating and facilitating the utilization of surface charges. Stronger electric field strength in the corresponding space-charge layer greatly enhances the separation efficiency of the photogenerated electron-hole pairs, ultimately enhancing their PEC performance. The synthesized $MoS_2/WS_2/Cu_2O$ exhibited a good electrical rectification and the best solar cell performance of0.58%higher conversion efficiency than that of theCu_2O0.036%sample.

Conflict of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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