

Unveiled Dual-function Antimony Selenide Nanowires for Self-powered Photodetector and Dye-sensitized Solar Cells

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ABSTRACT

Antimony selenide (Sb_2Se_3) attracted as a potential candidate as an absorber for photovoltaic application. Antimony selenide for self-powered photodetection and counter-electroded dye sensitized solar cell (DSSC) remnant unexplored much. Herein, device-grade nanowire Sb_2Se_3 film is developed through a solution process spray coat method, studied its light traction and solar cell application. The developed photodetector showed responsivity of $1.5 \mu\text{A/W}$, specific detectivity of 6.5×10^8 Jones, and ultra-fast response speed with rise time of 250 ms, decay times of 670 ms. Antimony selenide-based counter electrode based DSSC witnessed 4.18 % of power conversion efficiency.

Key words: Antimony selenide, Dual-function, Dye sensitized solar cell, Nanorods, Self-powered photodetector.

1. INTRODUCTION

The global energy requirement is increasing fast compared to the oil resources, research, and its advancement of alternate fossil fuel is high [1]. Photovoltaics technology, proved production of electric energy from light sources, is one of the promising renewable energy devices [2]. Numerous approaches, such as heterojunction solar cells, dye sensitized solar cells (DSSCs), and polymer and perovskite solar cell technology, have noticed their enormous growth. DSSC is a convenient, cost-effective energy conversion device, comprising a photoelectrode (PE), counter electrode (CE), and the electrolyte is sandwiched between PE and CE [3-5]. The CE plays a significant role as a multifunctional characteristic as it catalyzes dye redox shuttle and possesses wide chemical insistence, good conductivity for transfer of electron, and enhanced reflection nature, etc. [6].

Optoelectronic devices, which include photodetector (PD) or photosensor (PS), convert light signals to electrical signals, are essential in diverse applications of optical wireless communications, missile warning, night vision cameras, etc. [7]. PDs exhibit rapid response to light simulation and possess the ability for transient light detection. PDs are categorized into two types based on spectral sensitivity of selective (distinct-spectrum) and broadband (wide-spectrum) PDs [8]. Recent advancement in materials for PD has become potential. Significant progress has been evidenced in progress of advanced multifunction materials for fabricate novel optoelectronic devices [9]. Widespread research has been made to reconnoiter the application of antimony trichalcogenides, its alloys in microelectronic devices including solar cells, thin film transistors, resistive switching devices, and PD. In particular, Sb_2Se_3 is less explored as PD and photoelectrochemical cells. As a result, efficient multifunctional materials have been developed. Among them, antimony triselenide (Sb_2Se_3) established cost-effective, earth-abundant constituents [10]. Antimony selenide (Sb_2Se_3) arisen as a promising photovoltaic absorber material due to its suitable band gap of 1.0–1.3 eV, high adsorption coefficient ($>10^5 \text{ cm}^{-1}$ for visible light), good stability, and high tolerance to moisture, oxygen, and ultraviolet light, excellent carrier mobility ($\sim 20 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$), and broad spectral response

[11], stable in harsh conditions such as high voltages and temperatures. [12]. Moreover, it is composed of abundantly available elements [10].

Efforts have been made in the advancement of Sb_2Se_3 as an absorber material for photovoltaic applications [13-15]. In contrast, Panpan Sun and team members in the year 2016 developed thermal evaporation of antimony selenide as novel CE material for DSSCs application proved 6.47% power conversion efficiency (PCE) in combination with carbon nanoparticles [16]. To the best of the authors' knowledge, no other reports are available in the literature in this direction. Nevertheless, in the present work, Sb_2Se_3 nanowires are developed for the CEs of DSSC and PD applications. The obtained results indicate that the developed Sb_2Se_3 nanowires evidenced excellent performance for sensing and photovoltaic applications.

2. EXPERIMENTAL SECTION

2.1. Synthesis of Sb_2Se_3 Nanowires through Hydrothermal Process

Sb_2Se_3 powder was synthesized through slightly modifying the earlier reports [16-18], through low temperature processed hydrothermal method. Briefly, 1 mmol of antimony trichloride (SbCl_3) (Sigma-Aldrich, ACS reagent, 99.0%) and 1.5 mmol selenium (Se) powder (Sigma-Aldrich, 99.5%) powder were thoroughly mixed in deionized (D.I.) water and 60 mL of hydrazine monohydrate ($\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$) (Sigma-Aldrich, Reagent grade, 97%), stirred for 2 h. The mixture

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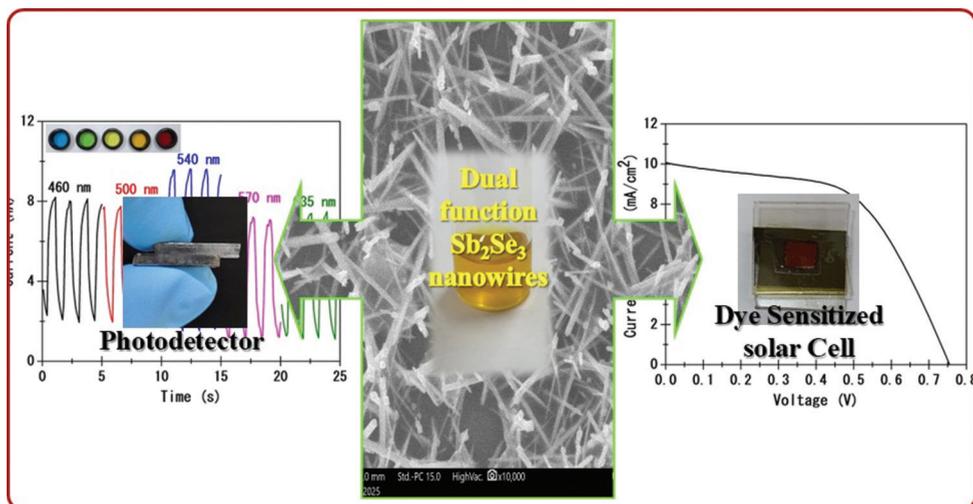
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GRAPHICAL ABSTRACT



solution was transferred into a Teflon-lined stainless steel autoclave and heated to 150°C for 18 h. After natural cool to 30°C, the precipitate was centrifugation, collected the precipitate and carefully rinsed with D.I. water, ethanol, and dried in an oven at 80°C for 6 h.

2.2. Development of Sb_2Se_3 -assisted PD

To fabricate the photoelectronic device, the synthesized Sb_2Se_3 powder (100 mg) was dissolved in a mixture of ethyl mercaptan (C_2H_5SH , 100 μ l) and 1,2-diaminoethane ($NH_2CH_2CH_2NH_2$, 1 mL) in a glass vial in ice bath condition, then sonication for 10 min, a clear homogeneous solution of yellow-brownish was obtained as shown in Figure 1a. The Sb_2Se_3 solution was sprayed over the FTO-coated glass substrate for 10 s, which is placed over a hotplate heated to 150°C. The obtained electrode is shown in Figure 1b. The resultant two similar substrates were sandwiched together by combining the Sb_2Se_3 film. The developed PD structure is similar to the reported work [17], and is Glass/FTO/ Sb_2Se_3 / Sb_2Se_3 /FTO/Glass, is shown in Figure 1c.

2.3. Fabrication of Spray-coated Sb_2Se_3 CE DSSC

The Sb_2Se_3 clear solution was spray-coated at 150°C on cleaned FTO-based glass substrates [Figure 2a], acting as CE. The standard PE was prepared as earlier reported [19,20], the PE structure was FTO/ TiO_2 /N719, and resultant substrate is shown in Figure 2b. The photo and CEs were sandwiched and the electrolyte solution. The Sb_2Se_3 -based fabricated DSSC is shown in Figure 2c.

2.4. Characterization

X-ray diffraction was carried out using PANalytical X'Pert3 X-ray diffractometer in between 0 and 60° of 2 θ value. The Sb_2Se_3 microstructure was examined through scanned using scanning electron microscopy (SEM) (JEOL, JSM-IT500), and the elemental analysis was made through EDAX studies. Optical absorption was done in 400–1000 nm wavelength region using of UV-Vis-NIR Spectrophotometer (Carry5000, Agilent Technologies). Raman spectroscopic study was made using HORIBA LabRam HR800 Raman Spectrometer.

The Current – Time ($I-t$) characteristics of the PD was measured using electrochemical workstation (IVIUMSTAT) at ambient condition. The Xenon lamp of 300 W equipped with optical filters was used as a light source to characterize the fabricated PD. The optical filters were used to achieve monochromatic light with different wavelengths (blue:

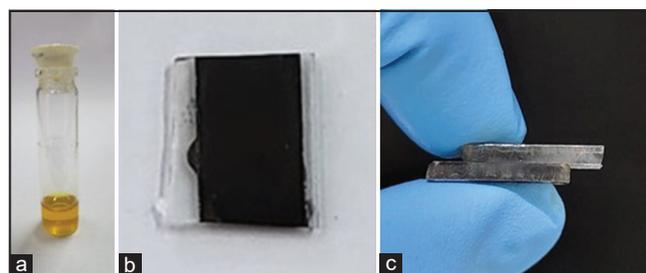


Figure 1: Sb_2Se_3 photodetector fabrication steps.

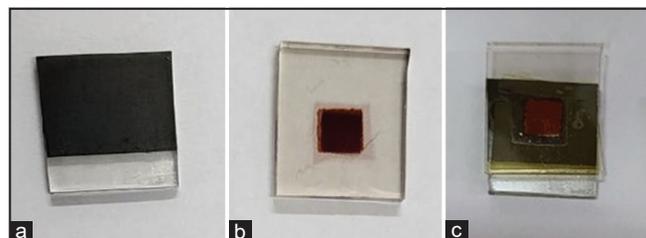


Figure 2: Spray coat Sb_2Se_3 based dye sensitized solar cell develop process.

460 nm, green: 500 nm, yellow: 540 nm, orange: 570 nm, red: 635 nm) from Xenon light. The Xenon lamp light intensity was varied by change in applied current to the lamp. The intensities of light powers of 28, 51, 76, 100 mW/cm^2 were used in the present investigation. The cyclic-voltammetry (C-V) measurement was made for the spray-coated Sb_2Se_3 electrodes, in three three-electrode system of Platinum wire as CE, Ag/AgCl electrode as reference, and Sb_2Se_3 -coated electrode act as working electrode. The 10 mM of LiI, 1 mM of I_2 , and 0.1 M $LiClO_4$ in acetonitrile-based electrolyte solution was used for C-V study. Tafel polarization was conducted to the symmetrical dummy test cells of two Sb_2Se_3 solution spray-coated counter electrodes sandwiched together, filled with I^- / I_3^- redox couple-based electrolyte solution. Whereas EIS study is conducted to the same symmetrical dummy test cells by apply 10 mV amplitude, in the frequency range of 0.1 Hz to 100 kHz. The photocurrent density - voltage (J-V) measurement for Sb_2Se_3 sprayed CE DSSCs performance was carried out with solar simulator (PEC-L01, PECCELL Inc., Japan, AM 1.5 filter attached) combined with a Keithley sourcemeter (2401N).

The intensity of light of the solar simulator was adjusted to 100 mW/cm^2 using calibrated mono-Si solar cell. The illumination mask with an aperture of 0.16 cm^2 was used during the J-V characteristics.

3. RESULTS AND DISCUSSION

The X-ray diffraction (XRD) pattern of the Sb_2Se_3 film is shown in Figure 3a, and results of all the diffraction peaks of as prepared film are well consistent with the crystalline orthorhombic Sb_2Se_3 (JCPDS # 75-1462) [15], and no existence of other phase or impurity, which directs its high-phase purity.

The spray-coated antimony triselenide film surface morphology is recorded through SEM, the resultant SEM image is depicted in Figure 3b. The results evidenced Sb_2Se_3 nanowires are uniformly distributed as similar to the earlier report [21]. Figure 3c shows the cross-section image of the resultant film. The elemental analysis of Sb_2Se_3 film is presented in Figure 3d, confirming the sample is good stoichiometric ratio (Sb of 48.25% and Se of 51.75%) as similar to the previous work [15]. The optical absorption of Sb_2Se_3 film is recorded, enabling their broad response to visible – NIR light, the resultant

broad, strong absorbance peak is present in the range of 400–850 nm, is shown in Figure 3e. The prepared film band gap energy (E_g) is derived from Tauc plot of $(ah\nu)^n = B(h\nu - E_g)$ ($n = 2$ for a direct bandgap semiconductor). The broad optical absorption observation is in good agreement with the reported study [22]. The bandgap evaluated for the prepared Sb_2Se_3 film, is 1.24 eV, and result permits its usage for solar cells. The narrow bandgap with broad absorption nature could enable the Sb_2Se_3 nanowires as a superior optically-active material for solar cells and PDs. The purity, surface oxidation of Sb_2Se_3 films are further assessed by Raman spectroscopy study, the revealed results are presented in Figure 3f. The Raman-active strong peak at 187 cm^{-1} and another weak peak at 209 cm^{-1} are assigned to Sb_2Se_3 [17,23,24] and peaks at 187 (strong), 250 (strong), 488 (weak) cm^{-1} are recognized as surface oxide Sb_2O_3 and the results are well matched with the reported studies [17,23,24].

The time-dependent photoresponse of the fabricated PD is recorded under zero bias condition by illuminate simulated white light of different powers $100, 76, 51, 28 \text{ mW/cm}^2$ and monochromatic visible light of 460 nm, 500 nm, 540 nm, 570 nm, 635 nm. The developed device is

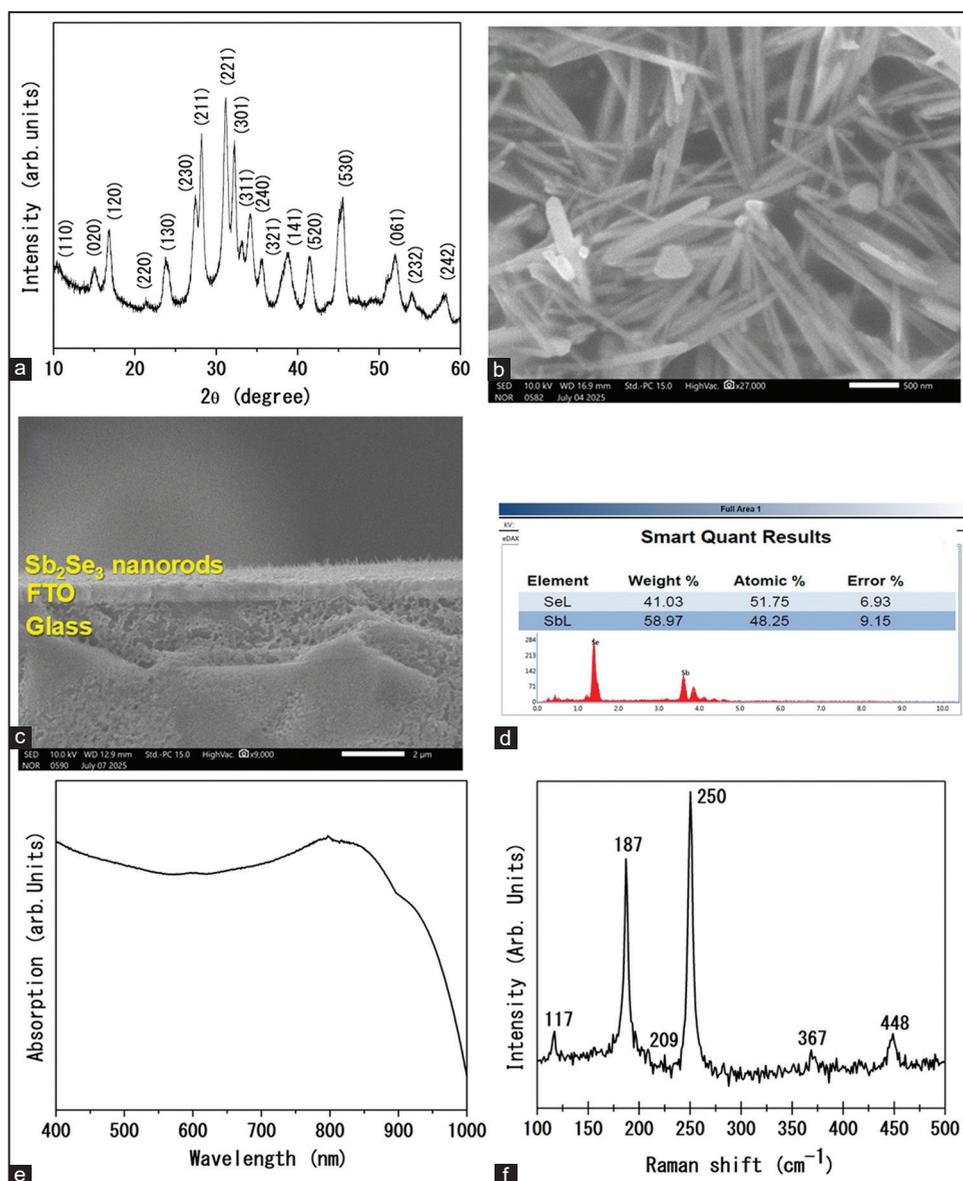


Figure 3: (a) X-ray diffraction, (b) surface morphology, (c) cross-section image, (d) EDAX results, (e) Optical absorption, (f) Raman spectrum of synthesized Sb_2Se_3 .

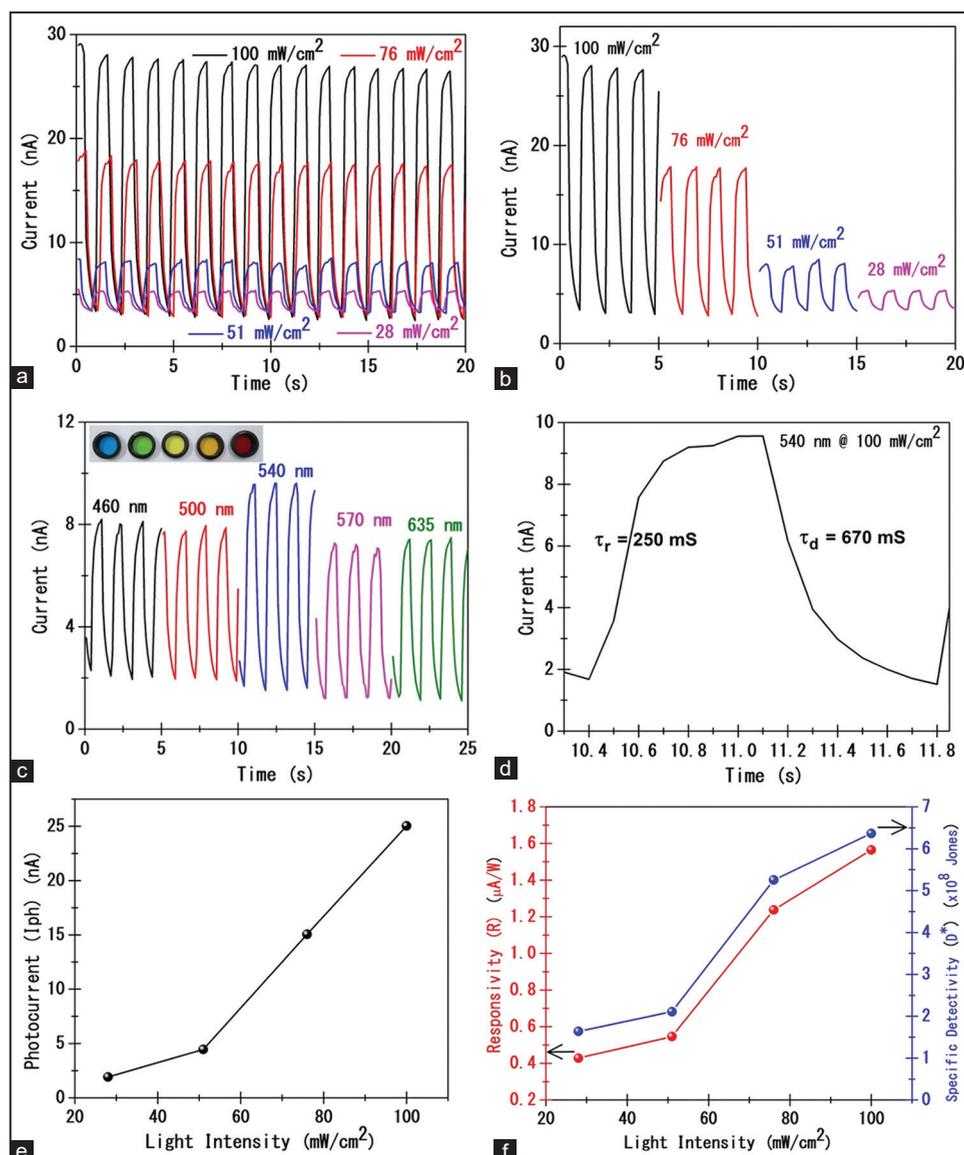


Figure 4: I-t curves for the sprayed Sb_2Se_3 photodetector.

shown as optically switching reliability without clear photocurrent decay after on and off cycles; the results are presented in Figure 4a and b. The on and off response is consistent with the absorption trend in the UV-Vis-NIR absorption spectrum. A on and off ratio ($I_{\text{ph}}/I_{\text{d}}$) of 9.2 at 100 mW/cm^2 discloses the good light detection ability of the developed sensor. It acted as a self-powered sensor that uses its own photovoltaic effect to sense the light at zero bias without any additional power. The photo response at different wavelengths of 460 nm, 500 nm, 540 nm, 570 nm, 635 nm of the prepared PD is shown in Figure 4c, and the light filters used in the present study are shown in Figure 4c. The rise and decay time response is presented in Figure 4d, in which the rise time of 250 ms and decay time of 670 ms are witnessed. The photocurrent response is directly proportional to the light intensity of the prepared PD, is shown in Figure 4e. The responsivity and specific detectivity response increase with light intensity trend is publicized in Figure 4f.

The C-V measurements are made in a three electrode system for evaluate electrocatalytic performance and its reversibility of the as prepared Sb_2Se_3 film in catalyze the reduction of I_3^- [25], the resultant C-V plot is presented in Figure 5a. As a result, two pairs of oxidation-reduction peaks are clearly observed in -0.4 – 1.2 V (vs. Ag/AgCl) range. The relative negative pair at low potential region is corresponds

to oxidation (Ox-1) and reduction (Red-1) of $\text{I}^- / \text{I}_3^-$, while positive pair at high potential region belongs to oxidation (Ox-2) and reduction (Red-2) of $\text{I}_2 / \text{I}_3^-$. Generally, CE serves to catalyze the reduction of I_3^- to I^- at the CE/electrolyte interface and are the very crucial parameters to estimate the catalytic performance of the CE [26]. The C-V results are presented in Figure 5a evidently shows the FTO- Sb_2Se_3 CE has a broad Red-1 peak with a current density of 0.72 mA/cm^2 , indicates the catalytic activity of Sb_2Se_3 CE is quite good.

To examine the charge transfer nature at catalyst-electrolyte interface of the prepared CEs, Tafel polarization and electrochemical impedance spectroscopic measurements are performed to the symmetric dummy cells. The symmetric dummy cells are assembled with two identical CEs (CE/electrolyte/CE). Figure 5b describes the Tafel polarization curve of symmetrical dummy cells, with $\log J$ as a function of voltage (V). The J_0 (exchange current density) is evaluated from the Tafel polarization curve to estimate catalytic performance of the CE. The J_0 is proportionate to the catalytic performance for I_3^- reduction and evaluated from the intersection of cathodic branch in Tafel zone with the equilibrium line, and is 1.5 mA/cm^2 . Figure 5c demonstrates a Nyquist plot of symmetric cells; its corresponding equivalent circuit is

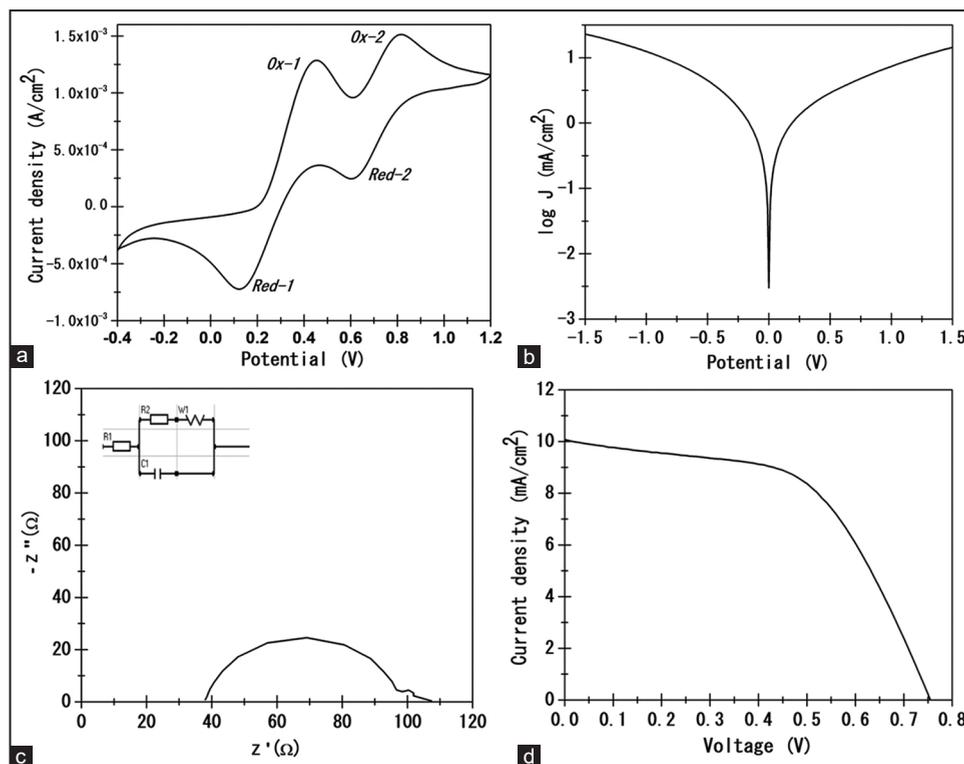


Figure 5: Sb_2Se_3 based (a) Cyclic-Voltammetry of counter electrode, (b) Tafel polarization, (c) electrochemical impedance spectroscopy performance of symmetrical dummy cell, (d) Current density – Voltage characteristics of dye sensitized solar cell.

shown in Figure 5c. The real (X-axis) intercept at higher frequency assigned to the series resistance (R_s) and is evaluated by fit the equivalent circuit, and is found 38Ω . The photovoltaic performance of the prepared CE Sb_2Se_3 is evaluated through current density – voltage (J-V) characteristics as shown in Figure 5d. Results demonstrated the PCE of DSSC made with Sb_2Se_3 is 4.18% with open-circuit voltage of 0.75 V, short-circuit density of 10.07 mA/cm^2 and fill factor of 0.55. The obtained results are comparable with the reported work of thermal evaporated Sb_2Se_3 CE DSSC [16].

4. CONCLUSIONS

In summary, the authors successfully synthesized the antimony selenide nanowires and prepared the device-quality films. The XRD, SEM, optical, optoelectronic properties, and Raman studies are thoroughly investigated. The PD and DSSCs are fabricated using Sb_2Se_3 nanowire-based electrodes. The fabricated PD shown R of $1.5 \mu\text{A/W}$ and D^* of 6.5×10^8 Jones and ultra-fast response speed with rise and decay times of 250/670 ms. The fabricated DSSC revealed PCE of 4.18%. The work carried out has quite potential for developing efficient multifunction materials for PD/sensors and energy conversion applications.

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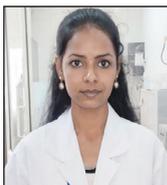
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***Bibliographical Sketch**

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Raghavender Mitty: Dr. Raghavender Mitty is a Physicist, interest research in Materials Science–photovoltaics, photo-rechargeable batteries and supercapacitor. He teaches post graduate Physics and Ph.D. course work. He is presently working as Associate Professor in the Department of Physics, Yogi Vemana University, India. He completed Ph.D. in Physics from Osmania University, India. He executed as Research Scientist at Aisin Cosmos R & D Ltd–Japan, IICT-India project. His present research work interests include fabrication of ambient photovoltaics, development and optimization of advanced solar cells, its integration with varies energy storage devices. Presently, he is working on photo-rechargeable batteries and supercapacitors, smart windows, so on.