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Zinc Oxide Nonmaterial Based Dye-Sensitized Solar Cells Using Natural Dyes Extracted from Different Plant Pigment

Wegene Lema Lachore

Faculty of Materials Science and Engineering, Jimma University, Jimma, Ethiopia

Email address:
wegekx@gmail.com

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Abstract: Dye-sensitized solar cells (DSSCs) are a low-cost alternative to thin-film and silicon-based solar cells. Platinum is an effective catalytic material for creating DSSC counter electrodes (CEs), but it is also pricey. In this work, PEDOT was used as a counter electrode and ZnO was used as a photoanode. Natural dyes such as Fragaria x ananass, Amaranthus Iresine Herbstii, Bougainvillea spectacles and flowers, and Beta vulgaris (beetroot) were extracted with four solvents to fabricate dye-sensitized solar cells (DSSCs). The zinc oxide (ZnO) nanoparticles were successfully synthesized by simple sol-gel techniques. X-ray diffraction (XRD) confirms that the synthesized materials were in the hexagonal crystal structure. The crystallite sizes of the ZnO NPs were found to be 29 nm. The optical properties of ZnO and extracted natural dyes were studied using UV-visible spectroscopy. The conversion efficiency of DSSC extracted from distilled water using Beta vulgaris sensitizers was found to be 0.0328%, which is good compared to the other natural dyes. For all sensitizers, the effects of several parameters were analyzed, such as incident photon switching efficiency (IPCE), short-circuit density (Jsc), fill factor, and open-circuit voltage (Voc).

Keywords: Dye-Sensitized Solar Cells, Natural Dye, Solvent, Electrolytes

1. Introduction

In today's world, energy is one of the most critical elements for daily existence. The majority of our current energy comes from nonrenewable sources, including charcoal, petroleum, and fossil fuels. Because of their ongoing CO_2 emissions, these energy sources have a negative influence on the environment, resulting in air pollution and climate change. To remedy this, researchers are striving to develop a low-cost, high-efficiency renewable energy source to replace fossil fuels, such as wind, hydropower, or solar energy. Solar cells are one of them, as they convert sunlight directly into electricity. Silicon-based solar cells, CIGS, CdTe, dye-sensitized solar cells, and quantum-dot solar cells are among the technologies used in solar cells. According to the energy sector, solar cells made of dye-sensitized solar cells (DSSCs) are being utilized to design and produce third-generation photovoltaic systems.

As a result, when compared to standard silicon-based solar cells, DSSCs are the most promising materials due to their better power conversion efficiency (PCE), cost-effectiveness, ease of manufacture, and environmental friendliness [1, 2]. As a result, DSSCs have been innovating in comparison to silicon-based solar cells. DSSCs have now achieved a worldwide efficiency of over 13%, making them viable options for producing clean and renewable energy [3]. DSSCs use dye molecules and a sensitive semiconductor electrode material to convert sunlight into electrical energy [4]. In DSSCs, metal oxide semiconductor nanomaterials like TiO_2, SnO_2, and ZnO are commonly employed as photoelectrodes [5]. Because of their greater potential applicability, ZnO nanostructures have been intensively studied for DSSCs [6–8]. Co-precipitation [9], hydrothermal method [10], sol-gel approach [11], microwave irradiation method [12], and chemical vapor deposition method [13] are some of the ways for producing ZnO nanostructures that have been reported in various publications. The sol-gel method for obtaining greater homogeneity in nanoparticle preparation is a simple, cost-effective, and low temperature method [11].

However, many studies using extracted natural dyes from natural sources and testing for DSSCs have lately been reported [14-16]. Karakus et al. used doctor-blade methods to fabricate DSSCs as sensitizers for pelagonium hortorum and pelargonium grandiflorum and TiO_2 as photoelectrodes and achieved a PCE of 0.065% and 0.067% [17]. Ramanarayanan et al. used red amaranth leaves and the effect of solvent to achieve a PCE of 0.23% and 0.530% [18]. In research conducted by Ammar, A. M., et al., DSSCs were built using dye derived from onion peels, red cabbage, and spinach leaves, with PCEs of 0.064723%, 0.060145%, and 0.171253% [19]. Khan et al. utilized N719 dyes with a Co and Ga co-doped ZnO thin film as an electrode to generate DSSCs with a PCE of 2.43% [11]. Sanjay et al. investigated ZnO nanorods manufactured by a hydrothermal method using dyes from Acalypha amentacea leaves and Peltophorum pterocarpum [10] with PCEs of 4.53% and 6.07%. To evaluate natural colors made from sambucus ebulus, Hosseinnezhad et al. employed solvents such as 1 N NaOH, 1 N HCl, and PCE [20]. Yirga et al. employed Bougainvillea spectabilis, Carissa Ovata, Hibiscus sabdariffa, Amarathus iresine herbisti, and Beta vulgaris for DSSCs with ethanol and water as solvents and ZnO as an electrode. PCE of 0.039% was obtained from Amarathus iresine herbisti [21].

Even though all of these studies found low PCE when compared to other conventional cells. The principles of operation and performance are still of interest, largely to create novel ways and get a deeper knowledge of the complicated cell.

In these works, we have reported our efforts in the synthesis of ZnO nanocolloids by the sol-gel method as photoanodes and Fragaria x ananass (strawberry fruit), Amaranthus Iresine Herbstii, Bougainvillea spectablis, and Beta vulgaris as sensitizers were extracted with solvents like ethanol, methanol, 0.1 M HCl, and distilled water for DSSCs. The crystalline structure and optical properties of the prepared ZnO nanocolloids were characterized. The optical absorption of dye extracts was studied using UV–Visible absorption spectroscopy, respectively. The J-V characterization was carried out to analyze the power conversion efficiency of the fabricated dye-sensitized solar cells.

2. Experimental Procedure

2.1. Synthesis of ZnO Nanoparticles

ZnO nanoparticles were synthesized by simple chemical the sol-gel method by using the precursor zinc acetate dehydrate for a zinc source and with potassium hydroxide used to adjust the pH of the solution [21]. In a typical experiment, 3.35mmol of Zinc acetate dihydrate ($Zn(CH_3CO_2)_2{\cdot}2H_2O$) was dissolved into 31.25 ml of methanol and the solution was vigorously stirred for 30 minutes with a magnetic stirrer. After 30 minutes, aqueous potassium hydroxide solution was added drop wise to the produced solution with steady magnetic stirring until the pH was close to nine and then stirred for 2 hrs at 60°C. After being aged for 24 hrs, the solution was centrifuged several times with distilled water and methanol to remove excess potassium ions. The precipitate was then dispersed in 12.5 ml of methanol and 2.5 ml of chloroform and dried overnight in an electric oven at oven at 80 OC. After calcining the produced samples at 600 OC for two hours, ZnO colloidal nanoparticles was obtained. These nanoparticles were used for further characterization.

2.2. Characterization Techniques

The structural analysis carried out by Shimadzu -7000 X- ray diffractomete with CuKα (λ= 1.5406 Å) radiation at room temperature. The optical properties of the samples were analyzed using an ultraviolet –visible spectroscopy in the wavelength rang of 200-800nm (Model: UV-3600 Plus).

An Electrochemical analyzer with computer control (Model- CHI-630A) without and with a monochromator, lamp horizon (Model 66182, Ser No, 227, MFD11/94) was used to analyze the Potential Vs Current (V-I) curve, and IPCE%. Denver instrument XE-50 to measure the mass of dye, drying materials (Compact 1300).

a) b) c) d)

Figure 1. *Extracted plants a) Fragaria x ananass b) Amaranthus Iresine c) Beta vulgarise and d) Bougainvillea spectablis.*

2.3. Extraction of Natural Dye

Figure 1 shows the samples of Fragaria x ananass (strawberry fruit), Amaranthus Iresine Herbstii, Bougainvillea spectablis, and Beta vulgaris (beetroot) were gathered and rinsed with running tap water to remove any dust particles. Then, for twenty-eight days at room temperature, a clean area was left in an open-air atmosphere. The samples are fully dried in a 70°C oven after twenty-eight days to remove any leftover moisture. The dried plant parts were then pulverized into powder and kept in plastic containers using micro-plant grinding equipment. For each plant, we used 1 gram of powdered plant in 35 ml of distilled water, ethanol, methanol, and distilled water under acidic conditions (0.1M HCl). Finally, the dye's absorption spectra were evaluated using UV-visible spectroscopy.

2.4. Preparation of Electrolyte

A homogeneous liquid electrolyte was generated by swirling 0.9 M of 1-ethyl-3-methyl imidazolium iodide (EMImI) with acetonitrile to make quasi solid-state electrolytes. To improve conductivity, 0.5 M sodium iodide was dissolved in the homogeneous liquid electrolyte above, then 0.12 M iodide and 3.5 percent w/w polyvinyl pyrrolidone were added (PVP). The liquid was heated to 70–80°C with vigorous stirring to dissolve the PVP polymer, and then cooled to room temperature to create a gel electrolyte [22]**.**

2.5. Fabrication of DSSCs

The indium tin oxide (ITO)-coated glass substrates were cleaned for 20 minutes using soap, distilled water, acetone, and ethanol, and then treated for 15 minutes with UV-O_3. To prepare the DSSC photoanode, the ZnO layer was combined with polyethylene glycol binder and formed onto the ITO-coated glass using the doctor blade procedure. To remove the adhesive tape, the produced ZnO film was air dried at 80°C for 20 minutes before being annealed at 400°C for 30 minutes. Overnight, the ZnO films were immersed in the dye solution. The dye film was washed with ethanol after the dye-sensitization process to remove the unanchored dye molecules before being employed to make the DSSCs. A PEDOT- coated ITO glass plate was used as the counter electrode. The dye- covered ZnO electrode and PEDOT counter electrode were constructed as a sandwich-type cell. The electrolyte solution was injected in between the cells which act as charge transport mediator between the ZnO photoanode and the PEDOT counter electrode in DSSC.

2.6. DSSC Assembly

The ZnO nanoparticles coated on ITO glass was placed facing upward, and the conductive side of the PEDOT-coated counter electrode faced the ZnO film. A DSSC was created by using capillary action to introduce liquid electrolyte (iodide/triiodide) into the gap between the ZnO film as photoanode and the counter electrode (cathode) as shown in Figure 2. To prevent the electrolyte from leaking, the two electrodes were clipped together with two binder's clips and bound together with mastics to create dye manufactured solar cells.

3. Result and Discussion

3.1. XRD Analysis

The prepared samples were analyzed by XRD using the CuKα wavelength of 0.15406 nm. Figure 3 shows the XRD result confirmed that the prepared samples was hexagonal crystal structure of ZnO nanoparticles with space group P63mc and lattice parameter a=b = 3.2498 Å, and c = 5.2066 Å and α = β = 90°C, γ = 120°C. The result is in a good agreement with the standard JCPDS card No. 036-1451. The incidence of sharp and number of diffraction peaks revealed the polycrystalline nature of the prepared nanomaterial.

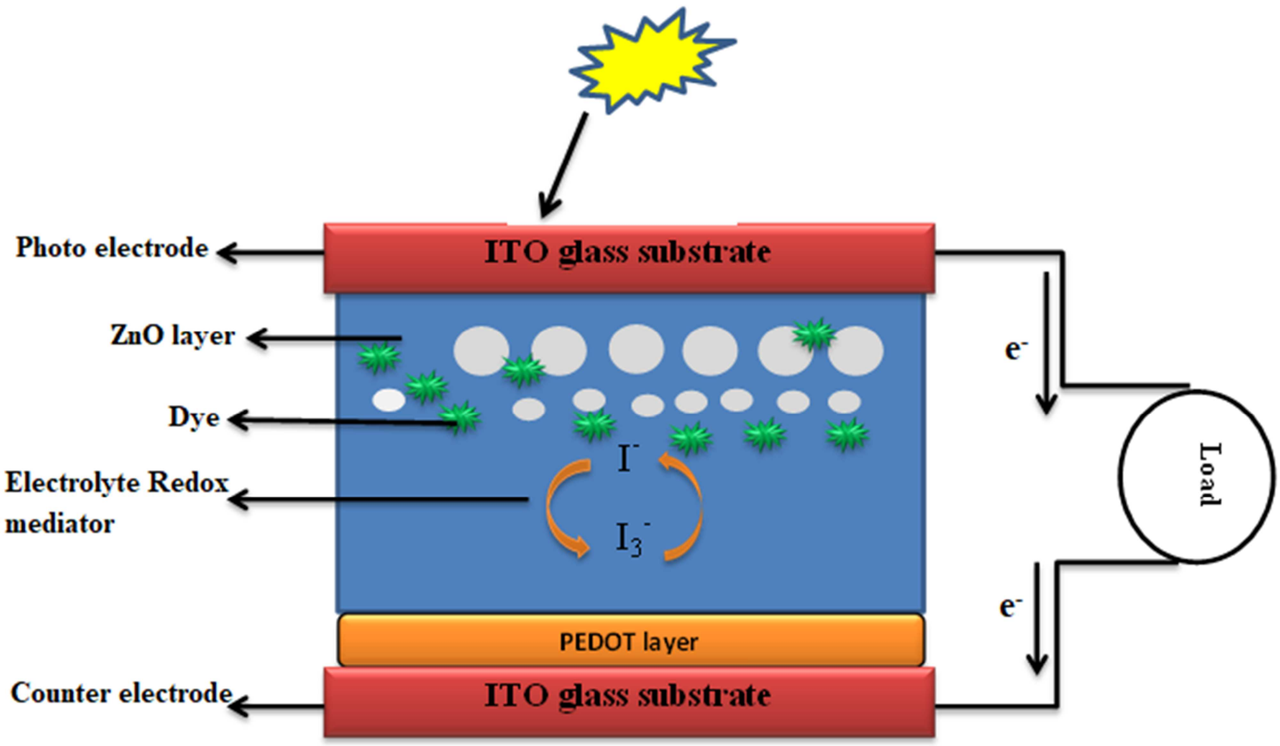

Figure 2. *Schematic diagram of DSSC assembly.*

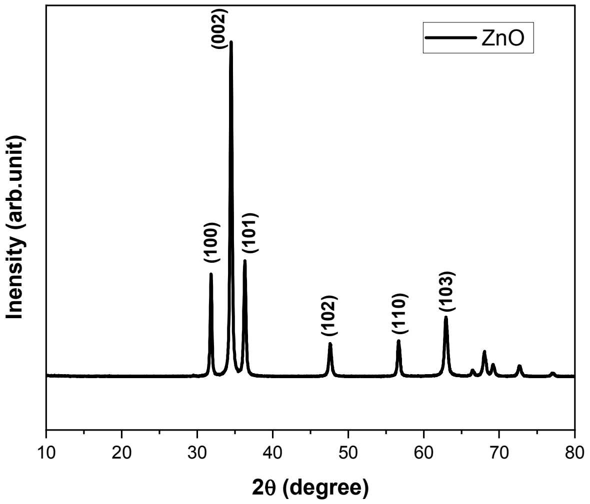

Figure 3. *XRD pattern of pure ZnO nanoparticles.*

The crystallite sizes for the prepared NPs were analyzed from the three most prominent diffraction peaks of (100), (002) and (101) planes using the Debye Scherer relation shown in equation 1 [23].

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (1)$$

Where, D is average crystallite size obtained from the three dominant peak, K is constant for the shape factor nearly equal to 0.9, λ is the wavelength of X-ray used (CuKα, 0.15406nm), β is the full width at half maximum and θ is the angel of diffraction of three most peak. The average crystallite size was calculated by using the (100), (002) and (101) peaks of ZnO NPs were found to 29 nm.

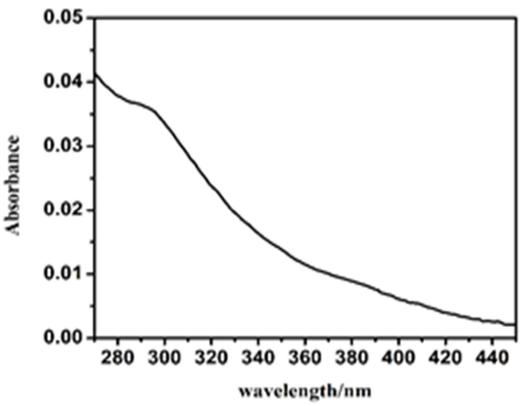

Figure 4. *UV-Vis absorption characteristics of ZnO nanoparticles dispersed in distilled water.*

3.2. UV–Vis Analysis

The UV-Visible spectroscopy measurements were taken to be 250-900 nm of ZnO nanoparticles. The absorption maxima were used to calculate the matching wavelength of 332 nm as shows in Figure 4. The sharpness of the absorption edge in single crystal ZnO is determined by the type of the electrical transition between the valence band and the conduction band [24].

The total performance of a DSSC is determined by the dye sensitizer's light absorption potential and the flow of the ejected electron through the ZnO layer. In this works four different types of natural dyes used for DSSCs. A UV-visible spectrometer model was used to measure the absorption spectra of the produced dye. Each sample extracted from ethanol (EtOH), methanol (MtOH), distilled water, and 0.1M HCl was measured for absorbance versus wavelength. The beta vulgaris dye extracts with 0.1 M HCl, water and ethanol exhibited the broad wavelength in the UV-Visible region of 539 nm (0.1M HCl), 477 nm-537 nm (water), and 475 nm (ethanol) and is shown in Figure 5a. This high absorption in the UV-visible range corresponds to chlorophyll-a and anthocyanin absorption. The absorption spectra of Amaranthus Iresine Herbstii dye extract from distilled water, 0.1 M HCl, methanol, and ethanol shows an intense broad absorption UV-Visible area of 532-675 nm (water), 535-673 nm (0.1M HCl), 416-668 nm (methanol), and 432 – 664 nm (ethanol) are shown in the Figure 5b. These indicating that the presence of anthocyanin in addition to chlorophyll-a, and chlorophyll-b. Figure 4c. shows the Bougainvillea leaf extracted from H_2O, 0.1M HCl, methanol, and ethanol. This dye had an absorption peak in the range of 533 nm, 537 nm, 665 nm, and 438-656 nm, which is the peak of anthocyanin and chlorophyll-b dyes. Figure 4a shows that there is no maximum absorption peak in the visible wavelength region in water extracts of Fragaria x ananass fruit. The absorption peak in the range of 667nm, 656nm, and 437- 667nm, which is the peak of chlorophyll-b containing dyes, can be seen in methanol, ethanol, and 0.1 MHCl extracts of Fragaria x ananass fruit shown in Figure 4d. The biggest peak absorption of wavelength occurs in the visible region at 380-400 nm.

The UV-visible data demonstrates that the extracts mostly contain betalains, chlorophyll, and anthocyanin, which the most remarkable feature is shown in all Figure 4.

The utilization of anthocyanins, which are found in the majority of plant components, as photosensitizers in the creation of DSSC has been extensively researched. Because its colour spans from red to blue in visible light, the anthocyanin molecule can be employed as a sensitizer for wide band gap semiconductors [25].

3.3. Photovoltaic Performance of DSSCs

As shown in Figures 5a-d Natural colours from Beta vulgaris (beetroot), Amaranthus iresine Herbstii, Bougainvillea spectablis, and flowers, Fragaria x ananass (strawberry fruit) with ZnO nanocolloid, were used to test the J-V properties of the produced DSSCs. The researchers have prepared devices that are extracted from ethanol and water. The PCE of the prepared DSSC from the water extract of Beta vulgaris was 0.033%, with a Voc of 0.27 V, a Jsc of 0.307 mA/cm^{-2}, and an FF of 0.396. This dye was also extracted with ethanol at 0.00504%, with Voc of 0.40, and Jsc of 0.043mA/cm^{-2} and 0.291. The DSSC prepared with Iresine Flower extracted by ethanol (EtOH) and water was 0.031%, Voc of 0.36 V, Jsc 0.25 mA/cm^{-2}, FF of 0.344 and 0.018%, Voc of 0.35V, Jsc 0.17 mA/cm^{-2}, and FF of 0.310. The DSSC device prepared from both Bougainvillea flower and Fragaria x ananass extracted with EtOH was 0.02%, Voc of 0.36V, Jsc 0.19 mA/cm^{-2}, FF of 0.29 and 0.025%, Voc of 0.37 V, Jsc 0.24 mA/cm^{-2}, and FF of 0.27. Among them, the DSSC constructed using ZnO sensitized by Beta vulgaris dye extract water had a better power conversion efficiency than the others. This is owing to the sensitizers' larger absorption spectrum, increased interaction between the ZnO nanocrystalline layer, and improved charge transfer. These results outperform those obtained in prior studies using DSSCs sensitized by various natural dyes [26, 27].

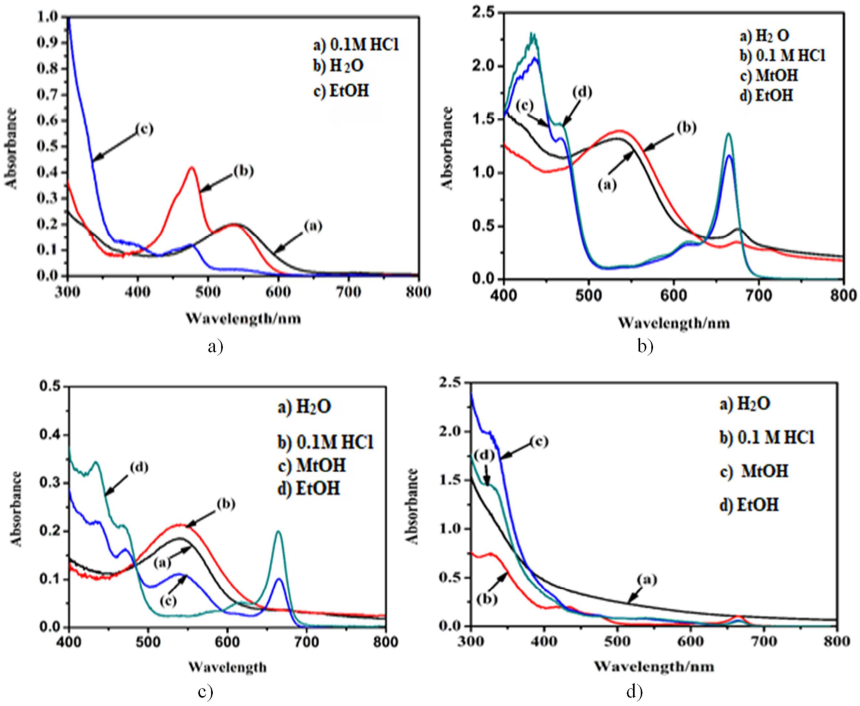

Figure 5. UV-visible spectrum of a) B. Vulgaris, b) Amaranthus Iresine Herbstii leaf, c) Bougainvillea leaf, d) Fragaria x ananass.

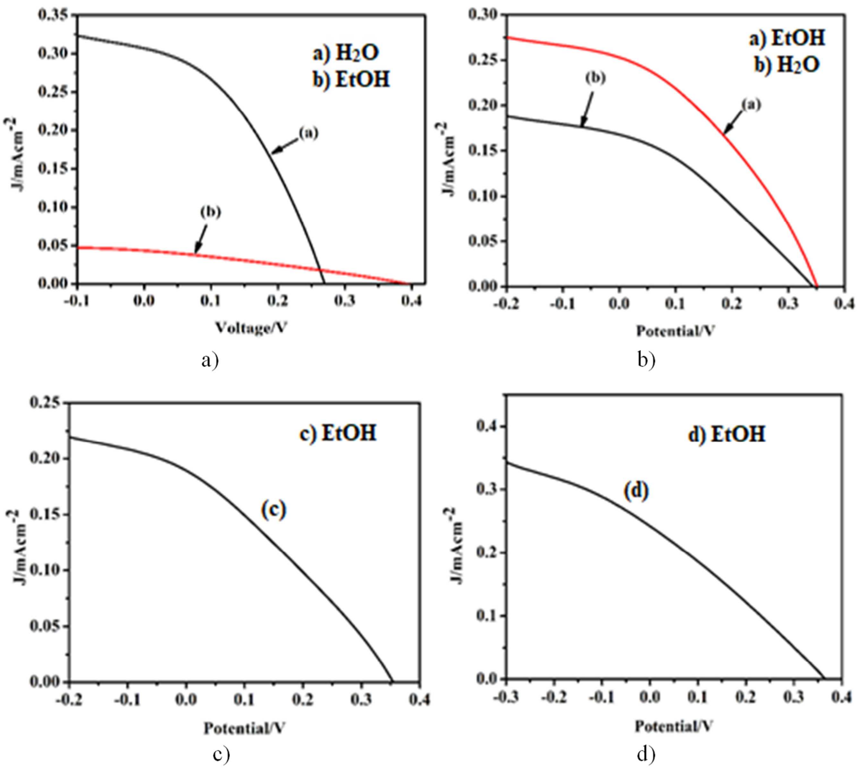

Figure 6. J-V of solar cell constructs using extracted dyes a) B. vulgaris, b) Amaranthus Iresine Herbstti leaf c) Bougainvillea leaf and d) Fragaria x ananass.

3.4. Incident Photon to Current Conversion Efficiency (IPCE)

The incident photon to current conversion efficiency (IPCE) of DSSC is defined as given by equation 2.

$$\mathrm{IPCE} = \frac{h\ c\ Jsc}{q\lambda\ Pin} \tag{2}$$

Where h Planck constant, c is the speed of light, λ is the wavelength of the incident light, and P_{in} is the intensity of the incident light and q is the elemntary charge. The incident photon to current conversion efficiency (IPCE) is defined as the number of photo-generated electrons in the external circuit divided by the number of incident photons at a given wavelength, and it provides another hint about the DSSCs' light harvesting performance. Figures 6a-d shows the IPCE spectra as a function of wavelength from 300 to 800nm for those four photoanodes. From IPCE result, fabricated DSSC with ZnO photoanode presents a maximum IPCE value of 77.1% at 350 nm for B. vulgaris distilled water extract and IPCE value of 66.4% at 340 nm for Fragaria x ananass ethanolic dye extract also IPCE value of 60.2% at 340 nm for Iresine flower distilled water extract. From overall Figure we have concluded that the photocurrent peaks occurring at approximately 360 nm, corresponding to energy of approximately 3.4 eV, are due to direct light harvesting by ZnO semiconductor, in which the photogenerated electrons diffuse through ZnO and the holes in the valence band are replenished by charge transfer from the I_3^-/I^- electrolyte.

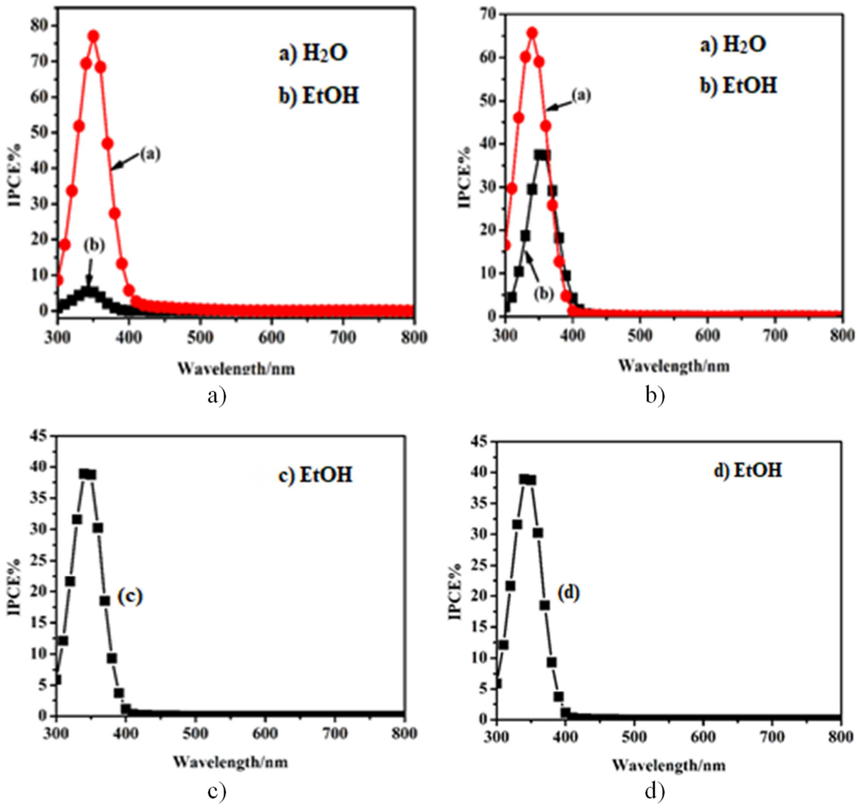

Figure 7. *IPCE curve of the fabricated DSSC based ZnO photoanode sensitizer extracted dyes a) B. vulgaris b) Amaranthus Iresine Herbstii leaf c) Fragaria x ananass d) Bougainvillea leaf.*

4. Conclusions

DSSCs are a low-cost photovoltaic technology in the field of renewable energy. In this work, four natural dyes were extracted with different solvents. To make dye-sensitized solar cells based on ZnO, natural dyes were extracted from B. vulgaris, Amaranthus iresine Herbstii, Bougainvillea leaf, and Fragaria x ananass with various solvents such as water, ethanol, methanol, and 0.1M HCl. The ZnO nanoparticles were synthesized using the sol-gel method. The XRD result confirmed that the prepared sample had a hexagonal crystal structure of ZnO nanoparticles. The average crystal size was found to be 29 nm. The absorption spectra and photoelectrochemical parameters of organic dyes from B. Vulgaris extracted with ethanol and distilled water, Bougainvillea leaf and Fragaria x ananass fruit extracted with ethanol, and Amaranthus Iresine Herbstii extracted with ethanol and distilled water were measured and characterized through UV–Vis spectrophotometry. The devices were prepared using natural dyes extracted from ethanol and distilled water. Among them, B. Vulgaris plant extracted in water based DSSC showed the best typical solar cell photocurrent density, photovoltage, fill factor, and overall efficiencies were JSC of 0.307 mA/cm^{-2}, VOC of 0.27 V, FF of 0.39571, and 0.033%. The efficiency for each dye is small because of the weak sanitization effect of the dyes, which results in small current and power conversion efficiency.

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