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RESEARCH PAPER

Graphene Sheets Incorporation in ZnO Nanostructure Thin Film for Enhancing the Performance of DSSC

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ABSTRACT

In the present work, the zinc oxide nanoparticle and its composite have prepared with the graphene as a photoanode electrode as well as synthesizing an excellent thin film from the PEDOT: PSS, which is conductive polymer loading with MWCNT as an auxiliary electrode for the DSSC. The photoanode characterized using (XRD, FTIR, Raman spectroscopy, BET-BJH, and UV-DRS). The FESEM and AFM performed to study morphology and structure of ZnO, ZnO/Graphene, and PEDOT: PSS/MWCNT thin films. The results confirm a successful fabrication of the thin films on the ITO by using the electrophoretic deposition method as well the added of graphene reduced the band gap close to 3.0eV. Moreover, the PEDOT: PSS/MWCNT nanocomposite based an auxiliary electrode offered high electrical conductivity, which enhanced the photovoltaic values of DSSC. The BET-BJH results demonstrated of the synthesis of zinc oxide nanoparticles at the surface area 6.66 m2/g and pore size 1.42 m^3/g.

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INTRODUCTION

Nanomaterials possess high applications, and they are widely attractive, especially in electronic devices [1]. ZnO nanoparticle has broad applications, including solar cells [2] antibacterial [3] electronic optics [1] battery [4] gas sensor and photocatalyst [5] self-cleaning coating [6]. The use of essential nanoparticles such as metal oxide depends on the shape, size, and condition of preparation method [7] .nanocomposites of metal oxide/graphene has many applications in nanotechnology of DSSC [8]. Considering graphene, it is one allotrope of carbon, which is a 2-dimensional and hexagonal lattice [9]. Attaching with metal oxide TiO_2 leads to ZnO improvement of a photoanode electrode in DSSCs and increasing efficiency of conversion to more than 12% [10]. TiO2/G film as a photoanode electrode with high porosity and surface area and resistance of electron is slow, and it has a high-frequency peak[11] more than TiO_2 nanoparticles. The surface area increases when graphene is adhesive on metal oxide or heavy metal Pd working as a retardant of aggregation of titanium nanoparticles [12] for improvement of semiconductor ZnO treatment with carbon nanoparticles to provide chemical and physical properties differently from ZnO nanoparticles [13]. Among applications of ZnO/Gr nanocomposites, are the use of these composites for adsorption of heavy metals such as (Cr, Cd, and Cu) from aqueous solutions [14] and drilling Oil and gas well [15]. ZnO/Gr nanocomposites have been employed in DSSC, and the obtained conversion efficiency was equal to 3.98 [16], whereas ZnO/GR nanocomposite obtained the conversion efficiency which was similar to 8.9[17].

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Fig. 1. Hydrothermal cell with a temperature Sensor.

Dye-sensitized solar cell considers the third generation of one types of solar cells that uses dye between its electrodes, which consists of four main components: a semiconductor electrode such as titanium oxide and nickel oxide, dye as a photosensitizer, electrolyte as an oxidative medium, as well as a counter electrode, which is often platinum or carbon [18] The type involves the use of photosensitizers for nanoparticles with different crystalline volumes. When the dye is combined with a semiconductor, it absorbs the ultraviolet light. It is useful in the sun, which results in an increase in the solar cell to more than 10% [19] The efficiency differs as the electrode is in the form of nanorods, Nanowires, Nano cons, and Nano leaves. These materials are used as electrodes after the coating on the base material (transparent glass conductor) [20]. Zinc oxide is used instead of titanium oxide. Studies showed that ZnO possesses many DSSCs [21], The dye or light sensor works by injecting the electron into the conduction band of the material of semiconductor, and then the loss of the electron from the photoanode electrode takes place to the auxiliary electrode in the DSSC solar cell [22]. The present work includes preparing zinc oxide nanostructure and incorporating the graphene sheets with ZnO. It fabricates the thin film on the ITO using the electrophoretic deposition method and investigates the DSSC.

MATERIALS AND METHODS

Materials and film characterization

The chemical materials which have been used in this study include, Znic acetate and polyphenyl pyrrolidine were purchased from Merck Co. Ltd,. N719 dye, PEDOT:PSS polymer and H_2PtCl_6 were purchased from Sigma Aldrich Co. Ltd,. But the material was purchased from supermarket company, USA and all of the chemical materials which have been used without further purification. The instruments which have been used in this study were (X-Ray diffraction "6000 XRD"), FT.I.R Spectrophotometer (Bruker 8400), Field-Emission Scanning electron, AFM (Atomic force microscope , AA4000, USA), Raman spectroscopy(Burker AXS Gmbh, Germany), DRS Spectroscopy (Quantachrome Instruments, USA), BET-BJH Surface Area Analyzer (Quantachrome Instruments, USA) for determination of pore size and volume and distribution of surface area, DC power supply (Phywe, Germany), 3465B digital multimeter, HP type, USA), and the PV power analyzer was supplied from SPD Co. Ltd to estimate the photocurrent-voltage parameters, Japan and used the Si solar cell to calibrate the intensity of incident light from the Xenon lamp (100 mW/cm^2) in simultaneously .

Preparation of photoanode and counter electrodes

Firstly, prepare zinc-oxide nanoparticles, A 2.3 mL solution of zinc acetate was prepared in ethanol. A solution of 2 g polyphenyl pyrrolidine was added in the distilled water, and the mixture was stirred for an hour. Then, a solution of potassium hydroxide was added to the mix and then was poured into the autoclave and was heated for 24 hours 80°C by using the homemade system to control temperature condition as shown in Fig. 1, then was cooled at room temperature

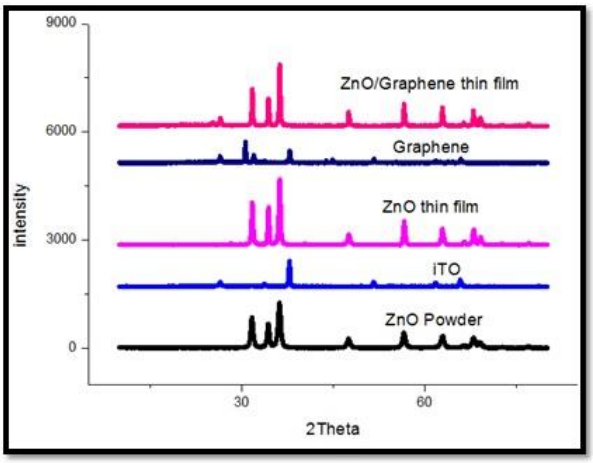

Fig. 2. XRD patterns of ZnO nanoparticles and ZnO/Graphene

and the resulting precipitation was filtered and was washed with ionic water and alcohol several times and then it was dried with 70°C in the air [23-25]. Zinc oxide graphene nanocomposite was prepared by dissolving 0.1 g of zinc oxide and, 0.03 g of graphene in 40 ml of ethanol and magnesium nitrate. Then the suspension was stirred in ultrasonic for three hours. The mixture was transferred to the electrophoretic container to deposit on the ITO glass. After that, the thin film was calcination 400°C for an hour. The polymer polyethene dioxythiophene PEDOT: PSS counter electrode was prepared. By took 10 mL of polymer and 0.03 g of MWCNT were dissolved in 30 ml of ethanol and then was transferred to the electrophoretic reaction vessel, and it was equipped with a 100-volt for 3 min to deposited on the ITO glass.

Assembly of the DSSC

Photoanode electrode was prepared by depositing both zinc oxide nanoparticles with its composites on the ITO conductive glass. The dye of the Ru complex prepared at a concentration of 1×10^{-4} molar in 20 ml of ethanol and acetonitrile. Then, the photoanode electrode was immersed in a dye solution for 24 hours in the dark at laboratory temperature, after washing with distilled water and ethanol and dry by the nitrogen flow. The electrolyte was prepared by adding some organic additives to increase the stability of the electrolyte 0.4 M of N-methyl-N-butyl-imidazolium iodide, 0.1M of lithium iodide and 0.04 M iodine in acetonitrile [26-28]. Photoanode and counter electrodes were clipped together after the injection of the electrolyte, and then the evaluation of parameters of the dye-sensitized solar cell was done

RESULTS AND DISCUSSION

According to the results of XRD patterns, the peaks were shown identical with the standard (JCPDS 36-1451), The film has appeared exhibit significant diffraction peaks at 31.760(100), 43.400(002), 36.230(101), 47.520(102), 56.580(103), 62.830(112), 67.920(201) which indicates the structural form of the zinc oxide nanoparticle a wurtzite hexagons. The results of the XRD spectra showed a value by 26.550 as identical peaks with a new miller index (002) of carbon atoms in the graphene and weaker than those shown in zinc oxide nanoparticle. This phenomenon due to the low graphene ratio compared to zinc oxide nanoparticle, which was equal to 25.8 nm, as shown in Fig. 2.

Moreover, the Raman spectrum of zinc oxide nanoparticle and its composite were recorded. Hexagonal wurtzite showed a sharp peak at 437cm^{-1}, that related to the zinc oxide nanoparticle of the E2 model. The two weak peaks at (331 and 580) cm^{-1} are attributed to A1 (TO) and (A1) LO [20-30] as shown in Fig. 3. While Raman spectra of ZnO/graphene showed a strong bond in 450 cm-1 and weak peaks in 150 cm-1 and 97 cm^{-1}, which results from the crystalline network of zinc oxide nanoparticle and packs 1320 cm-1 and 1538 cm^{-1} which results from the carbon atoms in the graphene as shown in Fig.3. Furthermore, the

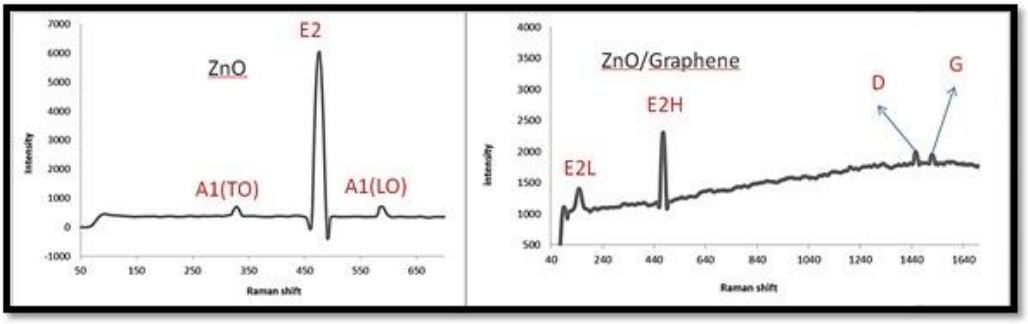

Fig.3. spectrum of Raman spectroscopy of the ZnO nanoparticles and ZnO/Graphene.

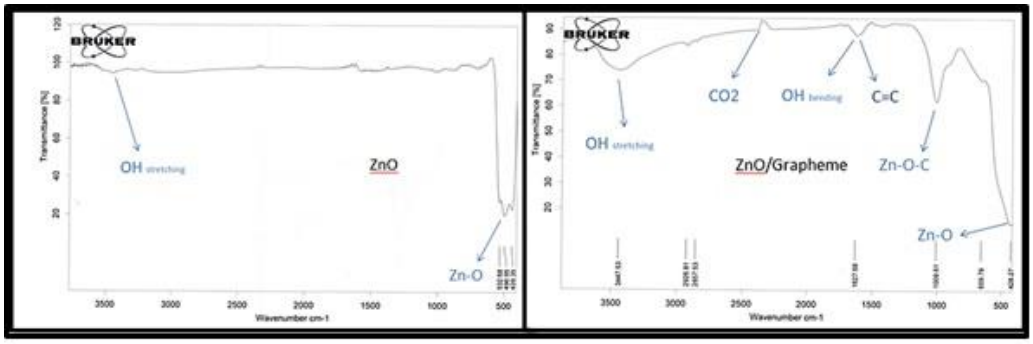

Fig. 4. FTIR of ZnO and ZnO/Graphene nanoparticles.

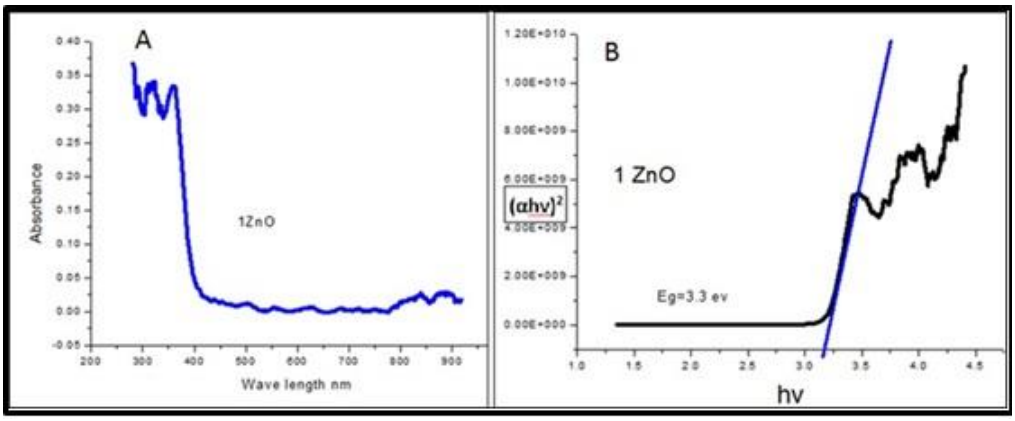

Fig.5. (A) DRS of ZnO nanoparticle (B) Band gap of ZnO nanoparticle

infrared measurements were taken for the ZnO nanoparticles and ZnO/ graphene composite, as illustrated in Fig.4.The results showed a broad and robust range of 450-100 cm^{-1}. The vibration of the metal-oxygen bond (Zn-O) appeared in two peaks positions; OH in 1620 cm-1are attributed to bending and at 3400 cm^{-1} due to adsorption of H_2O molecules on the surface of oxides [31-33]. While in ZnO/G composites C=C peak has appeared in 1620 cm^{-1} which often overlapped with OH peak.

Fig. 5 provides the spectrum of diffuse reflectance ultraviolet spectroscopy for zinc oxide nanoparticles was shown a wavelength of 375 nm. Thus, the energy band gap was equal to 3.3 eV. Whereas, the zinc oxide nanocomposite showed the wavelength of ultraviolet at 413 nm and the energy band gap equal to 3.0 eV showing a red shift compared to a band gap of ZnO nanoparticles, which is due to the decoration of zinc oxide by the graphene layer. The reason is related to the

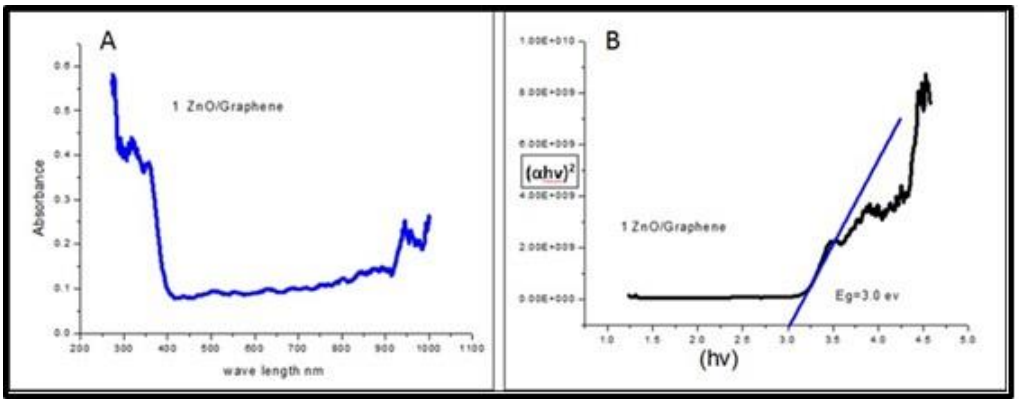

Fig. 6. (A) DRS of ZnO/G (B) Band gap of ZnO/G

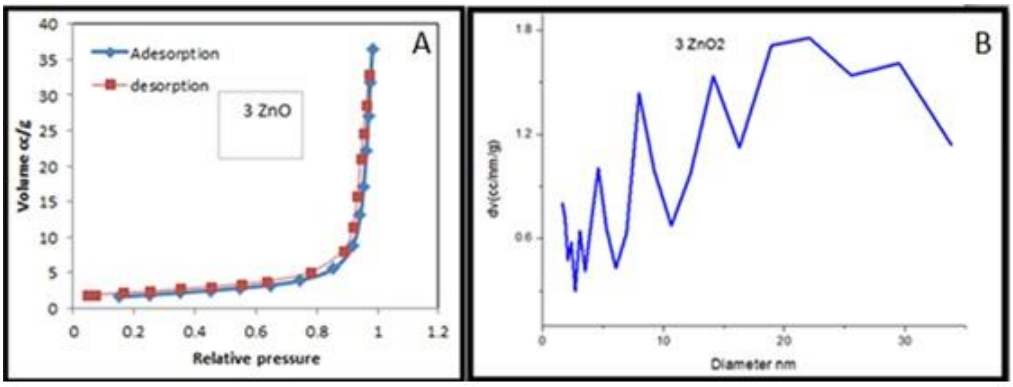

Fig. 7. (A) Adsorption-desorption isotherm (B) pore diameter of zinc oxide nanoparticles.

bonding between Zinc oxide and graphene, as shown in Fig. 6. The adsorption-desorption of ZnO nanoparticles with hysteresis loop (H3) at (P/Po = 0.2-0.9) was investigated. Indicating that the particles are in the form of (50 - 2) nm inter aggregations[34] as obtained by using the BJH and BET methods as depicted in Fig.7, as well the surface area equal to 6.66 m2/g, the pore size, and the pore diameter was equal to 1.42 m3/g and 22.70 nm, respectively.

Thus, the transmission electron microscopy was to investigate the crystalline structure of the surface and the shape, size, and distribution of the crystals, as exhibited from Fig.8. It shows a precise image of the ZnO nanoparticles, and the crystalline size of ZnO nanoparticles was also found. The high magnification of the ZnO nanoparticles' TEM image shows that it has a spherical shape small nanoparticle.

Also, to inspect and detect the surface morphologies and the structure of the electrodeposition of zinc oxide nanoparticle and ZnO/graphene and the thin films on the counter electrode surface, different methods as field emission electron microscopy and atomic force microscopy were used. From Fig. 9 (a,b), it

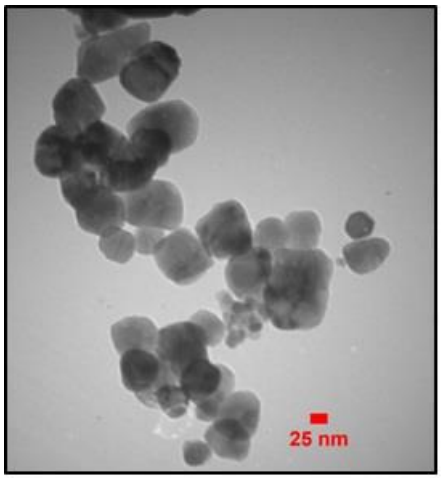

Fig. 8. TEM image of the zinc oxide nanoparticles.

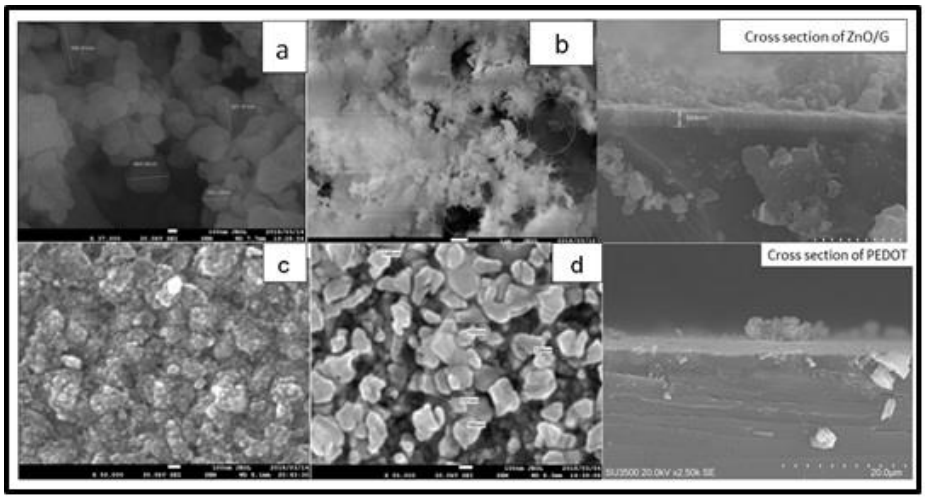

Fig. 9. FESEM images of (a) ZnO nanoparticles (b) ZnO/G nanocomposite (c) Platinum (d) PEDOT: PSS/MWCNT thin films.

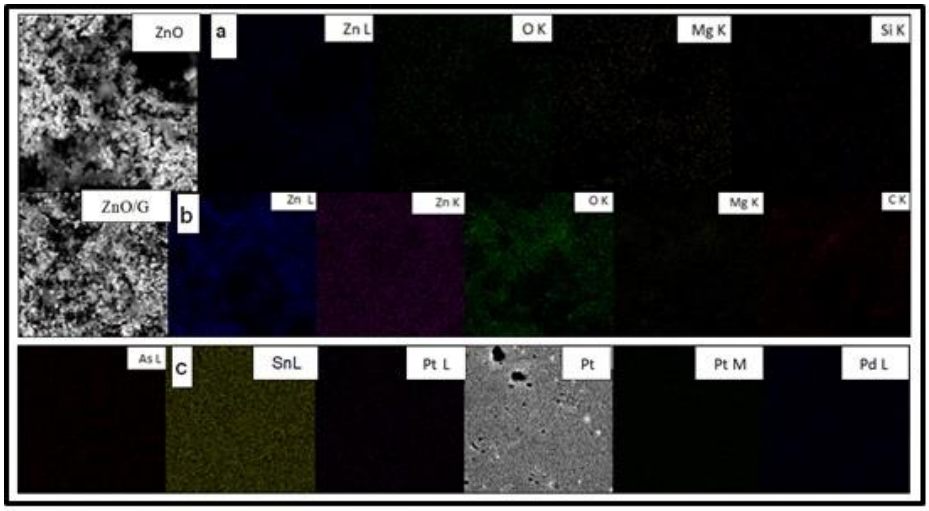

Fig. 10. Mapping images of (a) ZnO nanoparticles, (b) ZnO/G nanocomposite and (c) Platinum thin films.

appears the ZnO nanoparticles showed a spherical shape, which is consistent with the TEM result. In contrast, the ZnO/graphene image appeared ZnO nanoparticles were adhering to the graphene. This means the graphene appeared to be covered with ZnO nanoparticles and was more uniform with a thickness of about 884 nm, as shown from the cross-sectional image compared to other research results [35,36]. Fig. 9(c, d) exhibit evaluating the morphology of PEDOT: PSS/MWCNT and platinum electrodes. It appears the different geometric structure of PEDOT: PSS/MWCNT molecules (the size of particles was exceeded 100 nm) that an array on the counter surface and includes the molecules of MWCNT. Meanwhile, the high-order aggregate of platinum particles with small particles leads to the large active area for the auxiliary electrode.

The large active area would enhance electrical behavior via the facilitating charge-transfer process [12]. Mapping was used to evaluate the elements in the prepared nanoparticles. Mapping appeared a clear image with high dispersion of elements of all samples. The data confirm the samples' composition, which includes the main elements with the different energy-level shells. The existence of a trace of contaminants as silicon, magnesium, palladium, and tin from the precursors observed in mapping images as shown in Fig.10.

The atomic force microscopy of PEDOT also

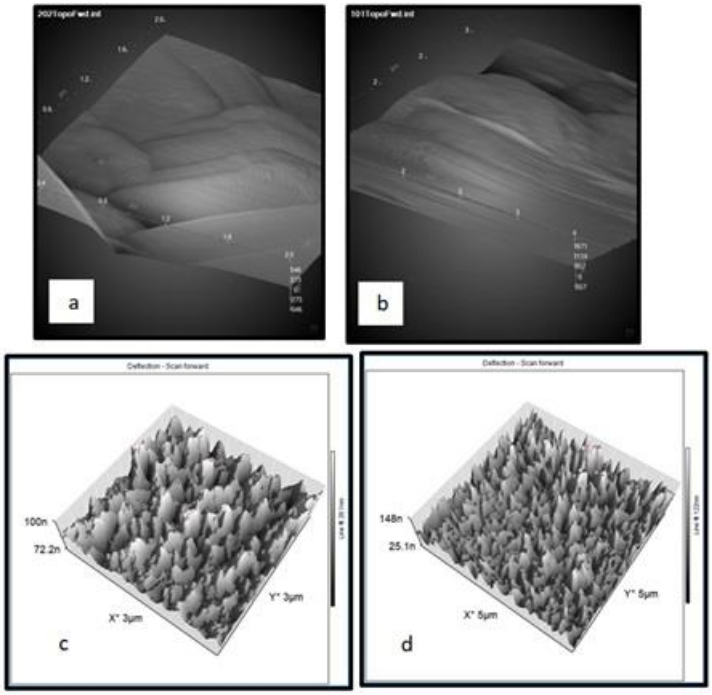

Fig. 11. AFM images of (a) ZnO nanoparticles, (b) ZnO/G nanocomposite, (c) Platinum, and (d) PEDOT: PSS/MWCNT thin films.

did the investigating of the surface morphology analysis: PSS/MWCNT and the platinum electrodes and the zinc oxide and ZnO/ graphene as photoanode for the dye-sensitized solar cell as presented in Fig. 11. The roughness parameters were assessed for the thin film of the zinc oxide and ZnO/ graphene electrodes such as (Ra and Rq) which confirmed that ZnO is smooth if compared with the ZnO nanocomposite that appeared significantly rougher. The variation in the values is remaining closed due to the added small amount from the graphene. So, Ra and Rq equal to 207.59 nm, 207.38 nm, and 252.30 nm, 274.08 nm for the ZnO and its nanocomposite, respectively. Also, it is noticed from the roughness value of the PEDOT: PSS/MWCNT thin film (Ra = 3.79 nm and Rq = 4.81 nm) and the platinum (R_a = 14.17 nm and R_q =17.01 nm) electrodes, as expected the values were four orders higher than that of the PEDOT: PSS/MWCNT thin film. It is clear from the results that the charge transfer resistance at the electrical double layer becomes lower in the platinum and PEDOT: PSS/MWCNT thin films. But, MWCNT thin film is high to enhance the electrolytic active site at the oxidation-reduction reactions [11].

Fig. 12. Illustrates the curve of photocurrent-voltage of the dye-sensitized solar cell with platinum, PEDOT: PSS/MWCNT, and MWCNT auxiliary electrodes, respectively. It is clear from the earlier results that incorporating the graphene layer with zinc oxide nanoparticles established the better properties for the thin film and consequently was select the nanocomposite electrode of the DSSC. Thus, the photovoltaic parameters such as

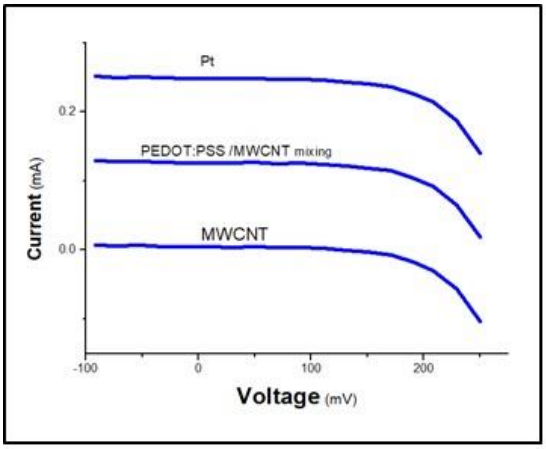

Fig.12. the photocurrent-voltage curves of ZnO nanocomposite with different an auxiliary electrodes of the dye-sensitized solar cell.

Table 1. Photovoltaic values of ZnO nanocomposite with different an auxiliary electrodes of the dye-sensitized solar cell.

Counter electrode	V_{oc}(mv)	J_{sc}(mA/cm^2)	V_{max}(mv)	I_{max}(mA)	FF	η
Pt	420	0.32	338	0.031	0.08	0.100
PEDOT: PSS /MWCNT	321	0.31	317	0.021	0.06	0.060
MWCNT	210	0.12	137	0.012	0.01	0.002

Voc, Jsc, fill factor, and the efficiency of the DSSC are tabulated in Table 1.

The photovoltaic parameters of the PEDOT: PSS/MWCNT electrode are higher if compared with the MWCNT electrode (concerning the platinum electrode as the main reference electrode). So, the η value of PEDOT: PSS/MWCNT composite film as an auxiliary electrode reaches 0.06, Which is improved from the MWCNT electrode. These results suggest that modification of MWCNT with PEDOT: PSS leads to enhance the η value.

The PEDOT: PSS/MWCNT counter electrode surface can provide and enhance the electro-catalytic process of the active site to complete the reduction of iodine ions (I_3^-/I^-) and improves the output of V_{oc} and in general the performance [37-40] of the work of the dye-sensitized solar cell. It is understood from data in Table 1, the efficiency of photovoltaic is lower, which indicates the recombination rate process[14,16] of the electron was fast as well as the thick of PEDOT: PSS/MWCNT may give a bigger resistance for the thin film. This result is consistent with the surface's morphology, as has been shown in FESEM and AFM images.

CONCLUSION

The present work includes synthesizing zinc oxide nanoparticles and its composite by incorporating the graphene layer with it. The XRD patterns refer to the wurtzite structure in the hexagonal model. Moreover, the Raman results enhanced the structure of the zinc oxide nanoparticle formed. The measurements of diffuse reflectance ultraviolet spectroscopy confirmed reduce band gap energy to become about 3.0 eV. The Brunauer-Emmett-Teller and Barrett- Joyner-Halenda methods were given information on the surface area and pore size, the values 6.66 m^2/g and 1.42 m^3/g, respectively. The thin film morphology of the surface electrode was inspected by the FESEM and AFM techniques. The roughness values that are calculated were enhanced the role of the counter electrodes' catalytic active sites. The photovoltaic parameters are estimated, and the results of Voc are high and got a low value of the efficiency of photovoltaic. This behavior because of the recombination rate of electron transfer.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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