Iranian Journal of Chemical Engineering

Journal Homepage: www.ijche.com

pISSN: 1735-5397
eISSN: 2008-2355

**Special Issue Article**

# Investigating the Performance of the Graphene-$WO_3$-$TiO_2$ Ternary Composite in Dye-Sensitized Solar Cells

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**ARTICLE INFO**

**Article history:**
*Received: 2024-01-09*
*Accepted: 2024-02-06*
*Available online: 2024-02-06*

**Keywords:**
*$TiO_2$,*
*$WO_3$,*
*Graphene,*
*DSSCs,*
*Nanostructures*

**ABSTRACT**

*A hydrothermal method was used to synthesize different photoanodes for their application in dye-sensitized solar cells (DSSC). These photoanodes included $WO_3$, $TiO_2$, Graphene-$TiO_2$, $WO_3$-$TiO_2$, and a nanostructure of Graphene-$WO_3$-$TiO_2$. The morphology of the nanoparticles was analyzed using the scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), ultraviolet-visible spectroscopy (UV-vis), and Fourier-transform infrared spectroscopy (FTIR). The results demonstrated that the graphene-$WO_3$-$TiO_2$ nanostructure had a large surface area, having provided more active sites for the efficient conversion of solar energy. Notably, the DSSC incorporating the graphene-$WO_3$-$TiO_2$ nanoparticle electrode outperformed cells based solely on $TiO_2$ and $WO_3$, achieving a higher short-circuit current density of 7.5 mA.cm$^{-2}$, an open-circuit voltage of 0.68 V, a fill factor of 0.46, and a power conversion efficiency of 2.4%. In contrast, the pure $TiO_2$ and $WO_3$ cells only achieved efficiencies of 0.88% and 0.69% Respectively. The excellent electron mobility of the ternary nanostructure facilitated the charge trapping and injection into the conductive substrate, reducing recombination. Additionally, the scattering effect of the $WO_3$ nanorods and graphene enhanced the light harvesting in the photoanode, leading to an increase in the overall efficiency of the solar cell. These findings highlight the potential of the synthesized graphene-$WO_3$-$TiO_2$ nanostructure as a promising photoanode material to be applied in DSSC.*

DOI: 10.22034/ijche.2024.434847.1512 URL: https://www.ijche.com/article_189842.html

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## 1. Introduction

Energy plays a crucial role in the economic development, population growth, and technological advancement[1]. However, the reliance on fossil fuels raises concerns about sustainability and future energy supplies[2]. To address these concerns, researchers have been exploring alternative energy sources, such as solar cells. Among the various types of solar cells, dye-sensitized solar cells (DSSCs) offer a promising solution due to their low cost and renewable nature[3, 4]. DSSCs are composed of porous nanocrystalline films sensitized from a dye, an electrolyte with a redox couple, and a platinized transparent conductive oxide (TCO)-coated glass substrate[5]. Titanium dioxide ($TiO_2$) is commonly used as the photoanode layer in DSSCs. $TiO_2$ enhances the efficiency of the solar cell by facilitating the electron transfer and providing a large surface area for the light and electron acceptance[6]. However, $TiO_2$ creates challenges due to its large energy bandgap, which hinders the electron transportation and fast charge recombination. These limitations restrict its effectiveness as a photoanode.[7].

To overcome these challenges and improve the efficiency of DSSC, researchers have proposed various strategies. One approach to address these challenges is doping $TiO_2$ with metals or non-metals. This can modify the electronic properties and bandgap of $TiO_2$, improving its performance as a photoanode[8]. Graphene is widely recognized as a remarkable material due to its high electron mobility, exceptional mechanical strength, and excellent thermal properties[9]. Muhamed et al. incorporated high-quality solvent-exfoliated graphene into transparent $TiO_2$, which led to a significant improvement in the power conversion efficiency (PCE) of DSSCs[10]. This improvement was achieved by reducing the recombination reaction rate and enhancing the electron transport within the cells[11]. It has been observed that a single layer of graphene absorbs approximately 1.2% of visible light. In the study by Chou et al., the impact of graphene on the scattering and electron transport properties of DSSCs was investigated. The authors discovered that the incorporation of graphene led to an increased PCE of the devices, attributed to the improvement in the electron transport and dye adsorption capabilities of the photoelectrode[12]. Shahid et al. indicated that doping graphene with $TiO_2$ increased the power conversion efficiency (PCE) by 33%[13]. Another approach involves pairing $TiO_2$ with a second semiconductor that has a lower bandgap energy. This heterojunction structure can enhance charge interaction and spatial separation of electrons, leading to the improved light absorption in the visible region and increased the efficiency of DSSC [14]. In recent studies, a heterogeneous structure of tungsten trioxide ($WO_3$) and $TiO_2$ has shown promises in improving the performance of DSSC. $WO_3$ exhibits a high absorption within the visible spectrum and low band gap energy, and long-term performance and perfect stability[15]. $WO_3$ nanostructures have relatively small band gap energies of approximately 2.2-2.9 eV, while $TiO_2$ mesoporous structures have high band gap energies of around 3.22 eV[16].

This structure reduces the bandgap of $TiO_2$, preventing charge carrier recombination and enabling a Z-scheme or heterojunction charge transfer pathway [17]. Yang et al. synthesized a $TiO_2$-$WO_3$ composite and reported a 20% increase in the efficiency of the solar cell using a non-platinum counter electrode[18].

It has been observed that the transfer of electrons from the conduction band of $TiO_2$ to the conduction band of $WO_3$ can minimize recombination losses. Additionally, the combination of semiconductor oxides with carbon-based materials such as Graphene can prolong the lifetime of electron-hole pairs and prevent their recombination[19]. Moreover, both graphene and $WO_3$ have photoinduced electron storage capabilities, making them suitable for the electron storage and mediation in DSSCs[20, 21].

In this study, we investigated the impact of graphene-$WO_3$-$TiO_2$ nanostructures on the PCE of DSSCs. To the best of our knowledge, there have been no previous reports on the utilization of a graphene-$WO_3$-$TiO_2$ ternary nanocomposite in DSSCs. By incorporating $WO_3$ and graphene into the $TiO_2$ layer, we aimed to enhance the overall performance of the solar cells. The introduction of $WO_3$ and graphene into the $TiO_2$ layer resulted in a reduction of surface trap states, effectively suppressing charge recombination. This improvement in charge transfer led to an increased driving force for the electron injection, consequently enhancing the power conversion efficiency of the solar cells. Our findings demonstrated that the solar cells with the Graphene-$WO_3$-$TiO_2$ composites as the photoanode exhibited significantly improved power conversion efficiencies compared to other cells. By exploring the potential of this novel composite, we hope to contribute to the development of more efficient and sustainable solar cell technologies.

## 2. Experimental

### *2.1. Materials and method*

To obtain the graphene-$WO_3$-$TiO_2$ nanostructure,14 miligrams of graphene (US research Nanomaterials, 2-18 nm with 32 layers) was sonicated in 150 mL of deionized water and subjected to ultrasonication for 5 minutes to achieve a graphene suspension. Then, 2 grams of $Na_2WO_3 \cdot 2H_2O$ (Merck) and 1 gram of NaCl (Merck) were dissolved in the graphene suspension. The mixture was stirred for 1 hour. The pH of the solution was carefully adjusted to approximately 2 by gradually adding drops of a 2.0 M HCl solution. Then, 1 gram of $TiO_2$ (US research Nanomaterials, anatase, 10-25 nm) was added to the solution and the solution was stirred for 3 hours. After preparing the dispersion, it was transferred into a Teflon-lined stainless steel autoclave with a capacity of 250 mL. The autoclave was then maintained at a temperature of 180 °C for a period of 24 hours. Following this, the autoclave was allowed to cool down gradually to reach room temperature. The synthesized product (graphene-$WO_3$-$TiO_2$ powder) was then centrifuged, washed with deionized water, and dried at 60 °C. For comparison, pure $TiO_2$,$WO_3$ , graphene-$TiO_2$ and $WO_3$-$TiO_2$ were also synthesized using the same procedure and conditions. The entire preparation process is illustrated in Figure 1. To prepare the paste for different photoanodes, 0.25 grams of semiconductors( $TiO_2$, Gr-$TiO_2$, $WO_3$-$TiO_2$, $WO_3$ and Gr-$WO_3$-$TiO_2$), 0.02 grams of Polyethylene glycol (PEG) and 1 ml of ethanol were combined. Then, the solution was stirred at a temperature of 60°C for 15 minutes. After that, 0.25 ml of Triton X-100 was added to the solution. Then, the solution was stirred for 3 hours.

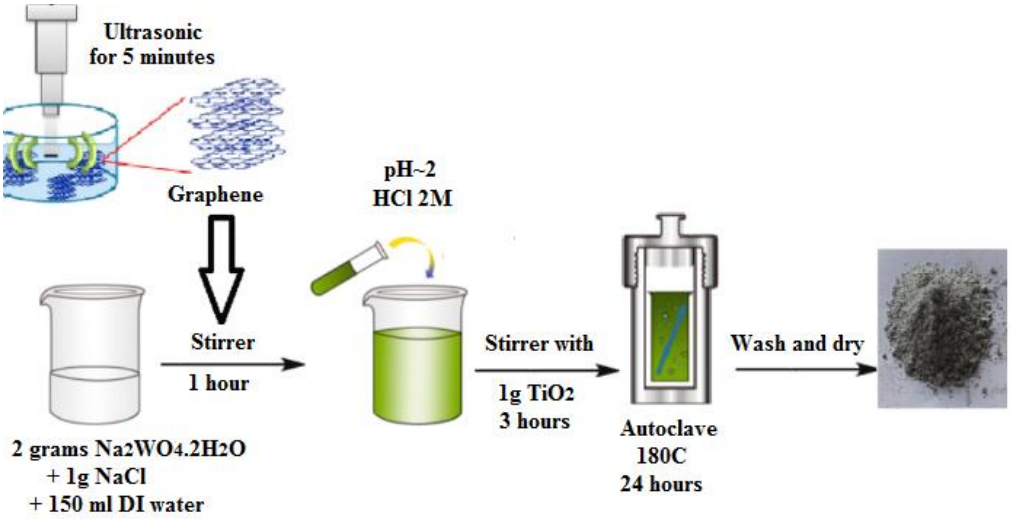


**Figure 1.** Schematic illustration of the process of synthesis for graphene-$WO_3$-$TiO_2$.

### *2.2. Fabrication of DSSC*

Thin films were prepared by applying a paste onto an ITO conducting glass plate using the doctor blade technique. The glass plate had a transmittance of 80% in the visible region. The prepared films were then annealed for 30 minutes at 450°C. To manufacture dye-sensitized solar cells, the thin film electrodes were submerged in a 0.1M N719 dye solution (Purchased from Sunlab) at room temperature for 24 hours. The electrodes were subsequently washed with anhydrous ethanol and dried. A platinum-coated ITO electrode was placed on top of the dye-absorbed electrode, and the cell's edges were sealed with a sealing sheet. Figure 2 illustrates the schematic representation of the dye-sensitized solar cells based on graphene-$WO_3$-$TiO_2$ thin films. For the redox electrolyte, a solvent consisting of the 0.5M KI, 0.05M $I_2$, and 0.5M 4-tertbutylpyridine was used. The surface morphology of the samples was examined using the scanning electron microscopy (SEMVEGA/TESCAN, Czech Republic). The FTIR spectrum was recorded in the range of 400-4000 $cm^{-1}$ using a Shimandzu FTIR spectrometer. The J-V curves of the DSSCs were measured using the Sharif solar I-V device with an AM 1.5G irradiation. The intensity of the incident light was calibrated to 100 mW $cm^{-2}$ using a standard silicon solar cell. The active areas of the cells were masked to 0.25 $cm^2$. The fill factor (FF) is calculated based on the J-V curve, where $FF = (J_{max} \times V_{max}) / (J_{sc} \times V_{oc})$. Here, $J_{max}$ and Vmax represent the photocurrent density and photovoltage at the maximum power output, while Jsc and Voc denote the short-circuit photocurrent density and open-circuit photovoltage respectively. Furthermore, the overall energy conversion efficiency (η) is defined as $\eta = (FF \times J_{sc} \times V_{oc}) / P_{in}$, where $P_{in}$ represents the power of the incident light.

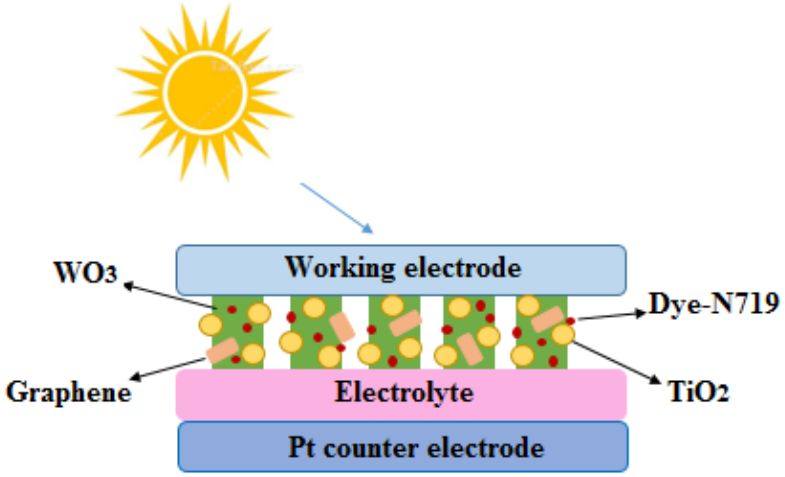


**Figure 2.** Schematic diagram of the graphene-$WO_3$-$TiO_2$ thin film based DSSC.

## *3. Results and discussion*

### *3.1. Structure and Morphology*

The morphology of $TiO_2$ and the graphene-$WO_3$-$TiO_2$ composite was examined using SEM images. In the image of pure $TiO_2$ (Figure-3a), the nanoparticles appear to be aggregated, which may affect the overall performance of the material. However, in the graphene-$WO_3$-$TiO_2$ composite (Figure-3b), the successful deposition of $TiO_2$ nanoparticles on the surface of 1D $WO_3$ nanorods can be observed, creating a strong connection between these two materials at the interface. This enhanced contact between $WO_3$ and $TiO_2$ can facilitate charge transfer during the photochemical reaction, which is beneficial for the overall performance of the composite[22]. Additionally, the SEM image reveals that the $WO_3$-$TiO_2$ composite exhibits a close contact, as $TiO_2$ nanoparticles grow on the surface of $WO_3$. This close contact between the two materials further promotes the efficient charge transfer. Moreover, it is evident that $WO_3$ contributes to the dispersion of $TiO_2$ nanoparticles, which can enhance the overall performance of the composite material[23]. Furthermore, the energy-dispersive X-ray spectroscopy (EDS) analysis (Figure-3c) confirms the successful combination of graphene, $WO_3$, and $TiO_2$. The EDS spectrum reveals the presence of five detected elements, including carbon (C), oxygen (O), tungsten (W), and titanium (Ti), further supporting the successful integration of these components in the graphene-$WO_3$-$TiO_2$ composite.These results provide important insights into the morphology and elemental composition of the $TiO_2$ and graphene-$WO_3$-$TiO_2$ composite, highlighting the successful combination of these materials and their potential for enhancing the performance of dye-sensitized solar cells.

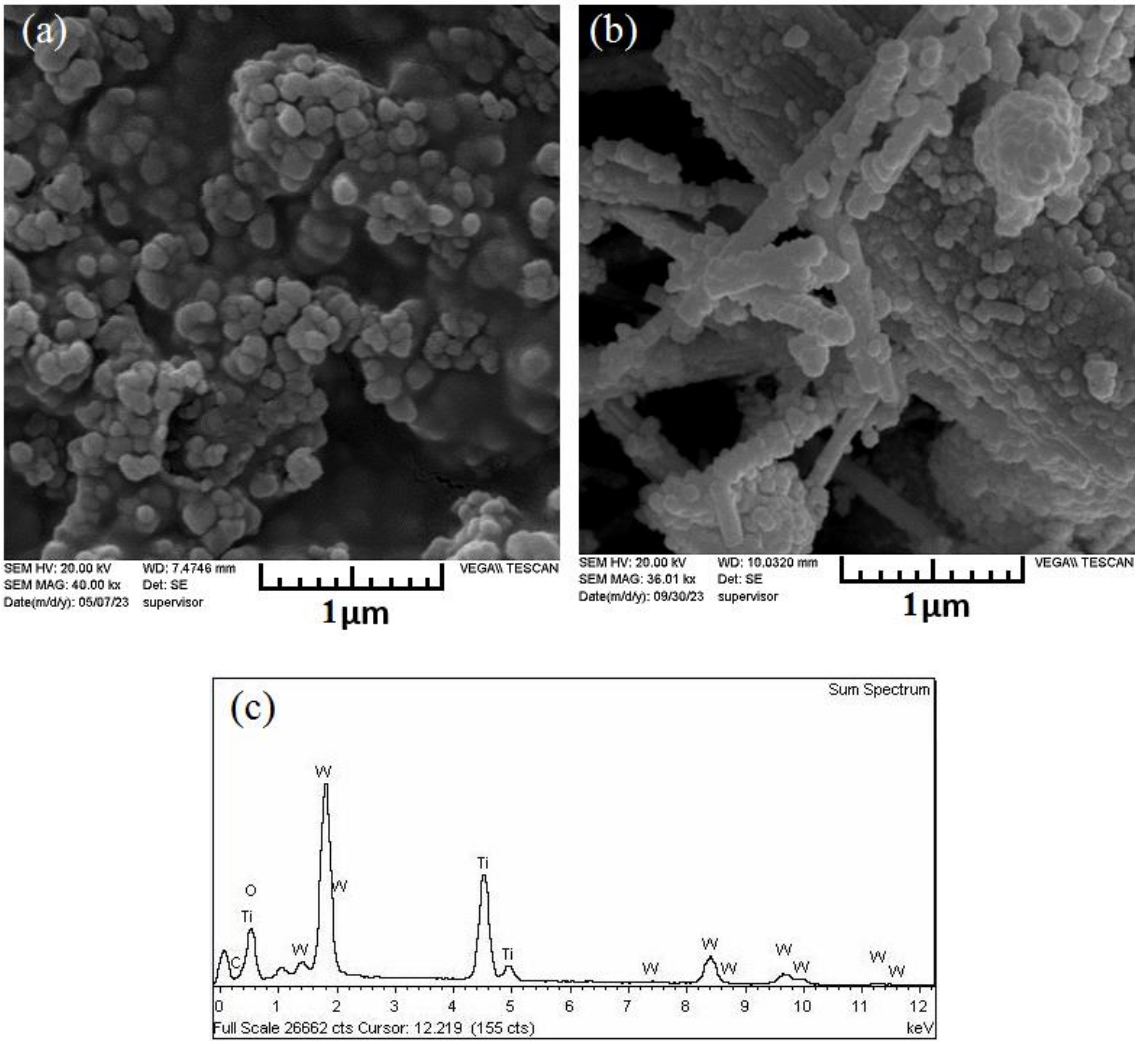


**Figure 3.** SEM image of the $TiO_2$ nanoparticle (a), SEM image of the graphene-$WO_3$-$TiO_2$ nanocomposite (b) and EDS spectra of graphene-$WO_3$-$TiO_2$ (c).

Figure 4 presents the EDX mapping of C, W, and Ti elements. The analysis of the $TiO_2$ sample, modified by $WO_3$ and graphene, revealed a notable concentration of W atoms, indicating the dispersion of W and carbon particles that formed aggregates. This suggests that $WO_3$ and carbon particles were well dispersed within the $TiO_2$ structure, ensuring a high and homogeneous distribution. The effective contact between these two semiconductors and graphene is crucial for facilitating the efficient electron transfer and enhancing the overall performance of the system.

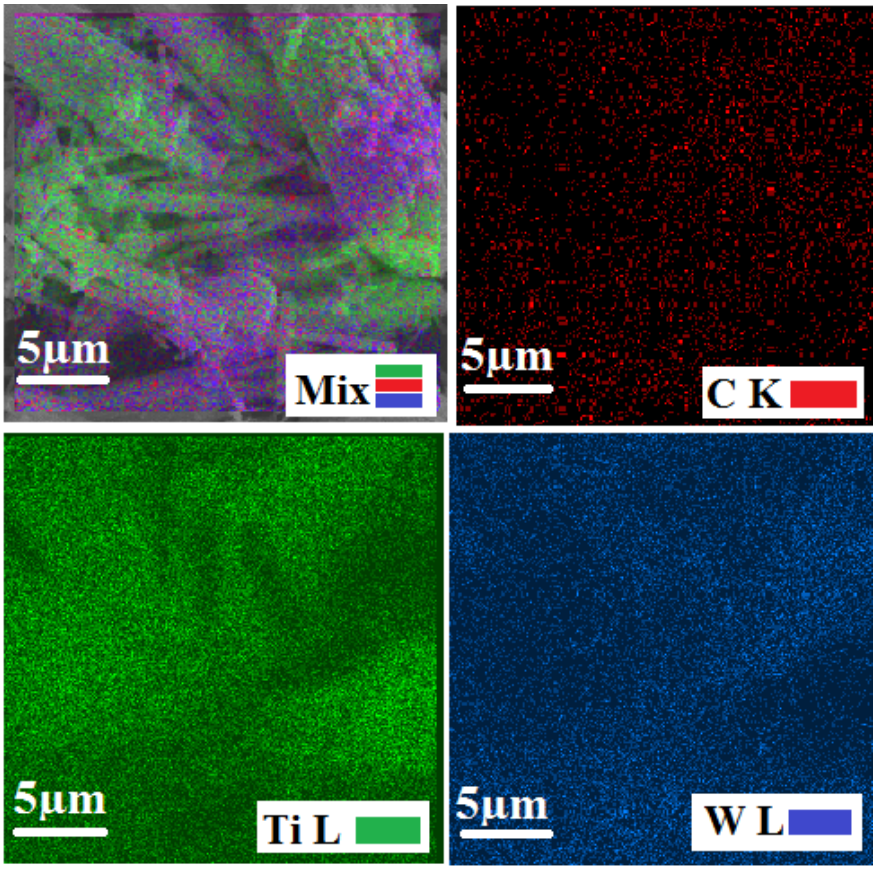


**Figure 4.** EDX mapping for the Graphene-$WO_3$-$TiO_2$ ternary nanostructure.

In Figure 5, the FTIR spectra of $TiO_2$, $WO_3$, and graphene-$WO_3$-$TiO_2$ are presented. Each sample exhibits a broad peak in the range of 3300-3600 $cm^{-1}$, which can be attributed to the stretching vibrations of -OH groups. Additionally, there are peaks observed in the spectra around 400-700 $cm^{-1}$, which can be assigned to the strong stretching vibrations of the Ti-O bond in the $TiO_2$ nanocrystal (indicated by the black curve)[24]. Furthermore, a peak at 820 $cm^{-1}$ corresponds to the vibration mode of the O-W-O bond in $WO_3$ (indicated by the red curve)[25]. Interestingly, the spectra of the graphene-$WO_3$-$TiO_2$ composite exhibit both absorption peaks, indicating the successful integration of $WO_3$ and $TiO_2$. This suggests that the composite material retains the distinct vibrational modes of both $WO_3$ and $TiO_2$, confirming their presence and interaction within the composite structure.

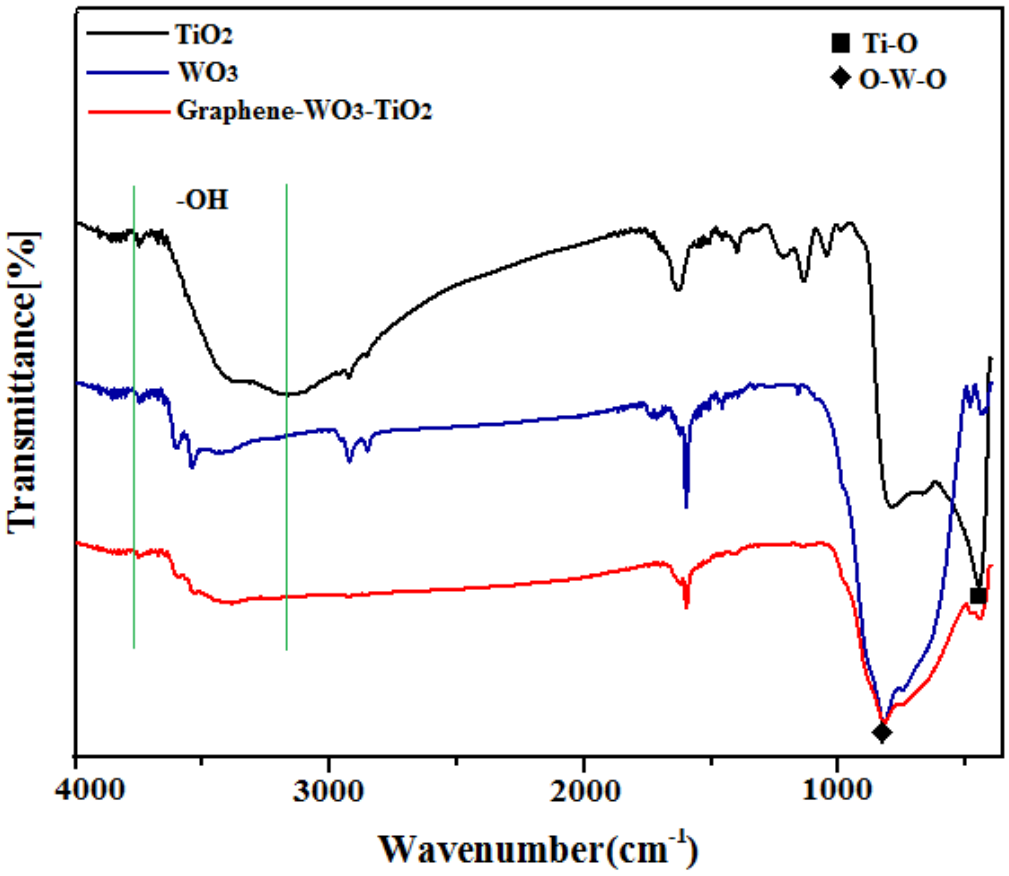


**Figure 5.** FTIR spectra of $TiO_2$, $WO_3$ and graphene-$WO_3$-$TiO_2$.

### 3.2. UV-VIS spectroscopy

The results presented in Figure 6 demonstrate the adsorption of N719 dye in a 0.1M NaOH solution on different photoanode films, including $TiO_2$, $WO_3$, Graphene-$TiO_2$, $WO_3$-$TiO_2$, and Graphene-$WO_3$-$TiO_2$. The absorption spectrum of the N719 dye was measured in the wavelength range of 450-750 nm when adsorbed on $TiO_2$ films with various compositions (Gr-$TiO_2$, $WO_3$-$TiO_2$, Gr-$WO_3$-$TiO_2$). Comparing the three nanocomposites (Gr-$TiO_2$, $WO_3$-$TiO_2$, and Gr-$WO_3$-$TiO_2$), Gr-$WO_3$-$TiO_2$ showed the highest absorbance,the amount of dye adsorption can be listed from the most to the least sequentially as: Gr-$WO_3$-$TiO_2$, Gr-$TiO_2$, $WO_3$-$TiO_2$, $TiO_2$, $WO_3$. Furthermore, the presence of graphene in $TiO_2$ led to the greater dye absorption compared to $WO_3$-$TiO_2$, indicating the enhanced dye adsorption properties of graphene. Additionally, the combination of all three materials resulted in the increased color absorption, thereby enhancing the efficiency of the solar cell.

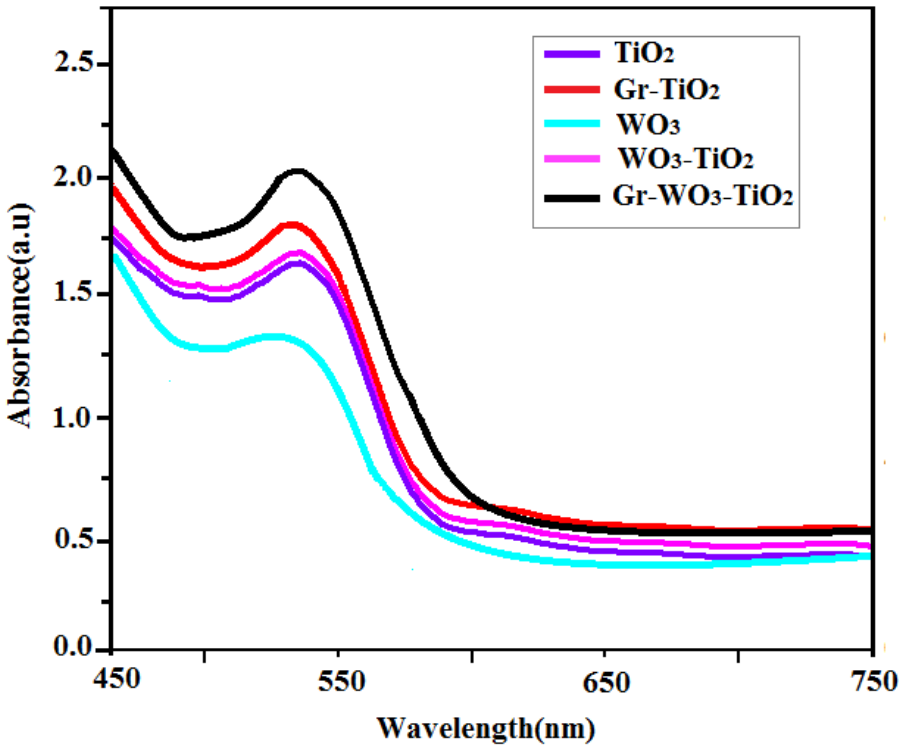


**Figure 6.** UV-Vis absorbance spectra of photoanodes.

### *3.2.Photovoltaic characterization*

As demonstrated in Figures 7(a) and 7(b), when DSSC exposed to solar radiation, the $TiO_2$ nanostructures captured electrons from the excited N719 dye molecules. These electrons then migrated to the conduction band (CB) of $WO_3$ due to the lower CB energy level of $WO_3$ compared to that of $TiO_2$. The solar cell efficiency of the Graphene-$WO_3$-$TiO_2$ photoanode was enhanced by the movement of holes ($h^+$) from the valence band (VB) of $WO_3$ to the valence band of $TiO_2$, facilitating the separation of electrons and holes. The lower band gap of $WO_3$ generated a photovoltaic effect in the high energy band gap of $TiO_2$ by promoting charge separation and extending the energy range of photo-excitation in the devices. Electrons in the Graphene, originating from the $WO_3$ CB, are rapidly transferred to the substrate without undergoing recombination. Zheng et al. conducted an investigation on the solar cell efficiency of a $WO_3$ photoanode-based. They performed the current density-voltage characterization and achieved a maximum efficiency of approximately 0.75% using a 12 μm thick $WO_3$ film[26].

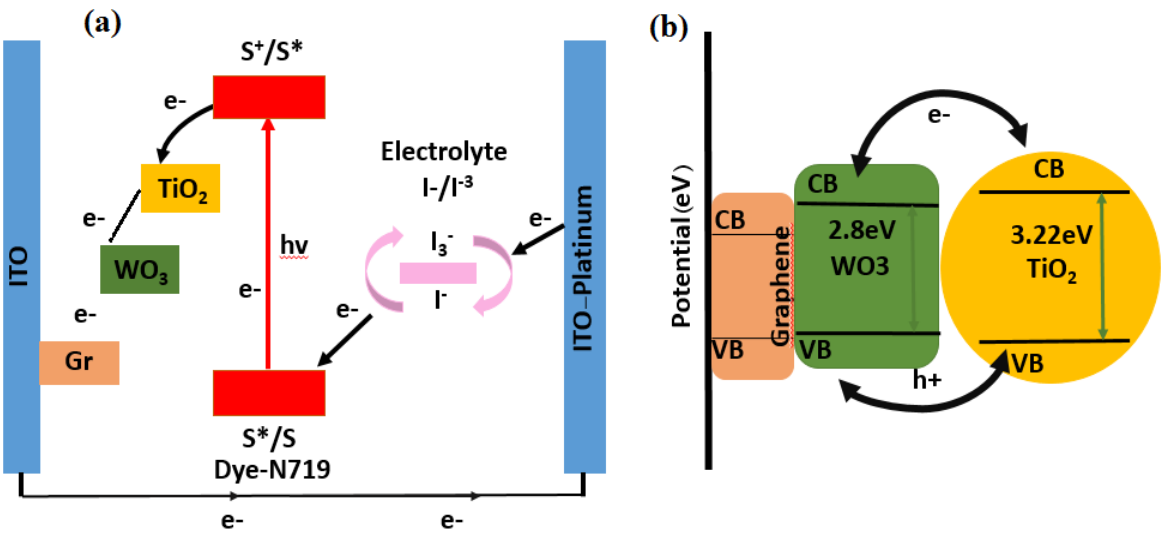


**Figure 7.** (a) Working mechanism of the DSSC with Graphene-$WO_3$-$TiO_2$ structures, (b) schematic diagrams of the electron transfer mechanism in the Graphene-$WO_3$-$TiO_2$ photoanode.

The J-V curves of the different DSSCs fabricated under the simulated 1.5 AM sunlight with front exposure are depicted in Figure 8. The photovoltaic parameters for the aforementioned DSSCs are presented in Table 1. The highest current density (Jsc) of 7.5 mA/cm$^2$ and open-circuit voltage (Voc) of 686 mV were achieved with DSSCs employing the ITO/graphene-$WO_3$-$TiO_2$ electrode.The best outcome within the mentioned parameters are overall power conversion efficiency (PCE) of 2.4% and a favorable fill factor of 46% for the $TiO_2$ modified with $WO_3$ and graphenecell. These results indicate a significant increase in Jsc, primarily attributed to the effects of the graphene and $TiO_2$ mixture. The improved open-circuit voltage observed in the composite electrode can be attributed to the enhanced electron lifetime in a one-dimensional electrode, allowing for the harvesting of more incident photons[18]. Furthermore, the presence of $WO_3$ in the composite electrode leads to a decrease in the internal resistance, confirming its capability to block charge recombination[27]. Table 1 demonstrates the positive effects of $WO_3$ on the Voc, Jsc, and PCE. In another experiment with a graphene-$TiO_2$ film, the corresponding DSSC exhibited a high light-to-electric energy conversion efficiency of 1.74%. It achieved a short-circuit current density of 5.96 mA/cm$^2$, open-circuit voltage of 645 mV, and fill factor of 45%. The appropriate content of graphene contributes to a more uniform distribution of $TiO_2$ nanoparticles, leading to an increased specific surface area of $TiO_2$ and enhanced dye adsorption. These factors contribute to improved Jsc and PCE. Overall, these results highlight the significance of incorporating graphene, $WO_3$, and $TiO_2$ in DSSCs, as they lead to the improved photovoltaic performance through the increased Jsc, enhanced open-circuit voltage, and optimized overall conversion efficiency.

**Table 1.** $J_{SC}$, $V_{OC}$, FF, and PCE of DSSCs fabricated using pure $TiO_2$ particles,$WO_3$, graphene-$TiO_2$ , $WO_3$-$TiO_2$ and ternary graphene-$WO_3$-$TiO_2$ nanostructure.

| Sample | Jsc ($mA/cm^2$) | Voc (mV) | FF % | PCE % |
|---|---|---|---|---|
| $TiO_2$ | 3.21 | 530 | 51 | 0.88 |
| $WO_3$ | 2.18 | 667 | 48 | 0.69 |
| Gr-$TiO_2$ | 5.96 | 645 | 45 | 1.74 |
| $WO_3$-$TiO_2$ | 4.462 | 548 | 69 | 1.68 |
| Gr-$TiO_2$-$WO_3$ | 7.5 | 686 | 46 | 2.4 |

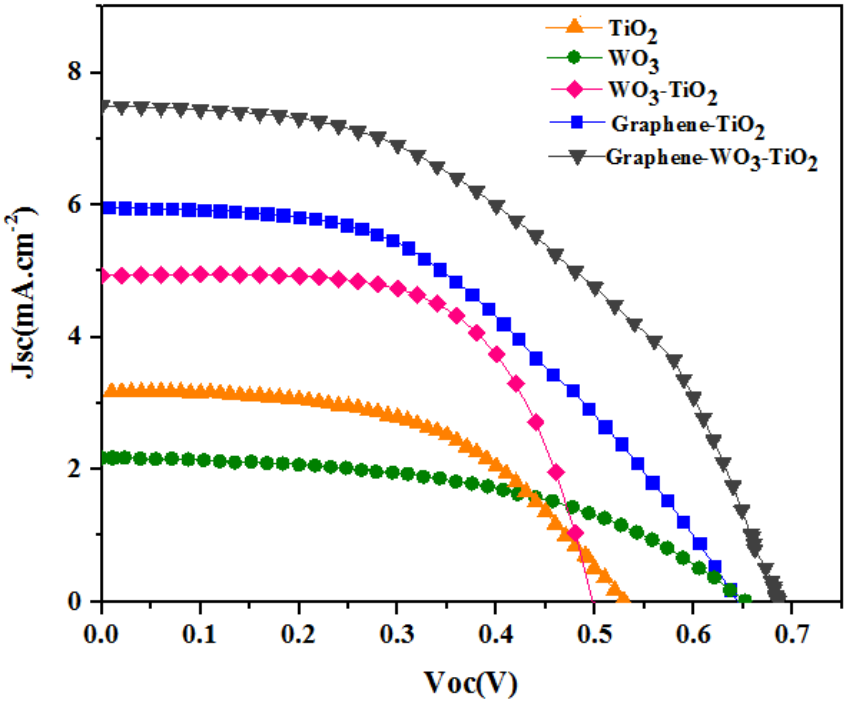


**Figure 8.** J-V characteristics of the DSSC with pure$TiO_2$, $WO_3$, graphene-$TiO_2$ , $WO_3$-$TiO_2$ and graphene-$WO_3$-$TiO_2$ photoanodes under the simulated 1.5 AM sunlight

### *4.Conclusion*

In this study, we successfully synthesized a nanostructure graphene-$WO_3$-$TiO_2$ using a simple hydrothermal method. The main aim was to investigate how this nanostructure affected the performance of dye-sensitized solar cells (DSSCs). The results showed that the graphene-WO3-TiO2 nanostructure, with its large surface area, was able to provide more reactive sites for the efficient solar energy conversion. The growth of Graphene-$WO_3$ nanostructures on the $TiO_2$ surface were found to be crucial in improving the solar efficiency of standard DSSCs. The alignment of the work

functions between $TiO_2$ and $WO_3$ allowed for the effective charge separation and reduced recombination of electron-hole pairs. The graphene-$WO_3$-$TiO_2$ photoanode film demonstrated superior performance compared to other nanostructures, thanks to the efficient electron trapping capabilities of Graphene and $WO_3$ nanostructures, which reduced the recombination rate of electron-hole pairs. To compare the photoanodes, $TiO_2$, $WO_3$, Graphene-$TiO_2$, and Graphene-$WO_3$-$TiO_2$ were treated hydrothermally at 180°C. The DSSCs fabricated with TiO2 showed the short-circuit current densities (Jsc) of 3.21 mA/cm$^2$, open-circuit potentials (Voc) of 0.53 V, and cell conversion efficiencies of 0.88%. The DSSCs prepared with $WO_3$ exhibited the Jscs of 2.18 mA/cm$^2$, Vocs of 0.66 V, and cell conversion efficiencies of 0.69%. However, when the Graphene-$WO_3$-$TiO_2$ composite was used, the DSSCs showed significant improvement, with the Jsc of 7.5 mA/cm2, Vocs of 0.68 V, and cell conversion efficiencies of 2.4%. These findings clearly demonstrate the remarkable enhancement in the photovoltaic performance achieved by incorporating the Graphene-$WO_3$-$TiO_2$ composite in DSSCs. The use of this composite material has proven to be highly beneficial, resulting in the increased short-circuit current density, open-circuit potential, and overall cell conversion efficiency.

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