



# Investigation of the Effect of rGO/TiO<sub>2</sub> on Photovoltaic Performance of DSSCs Devices

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# ABSTRACT

n this paper, four indoline-based organic dyes were introduced and studied as photosensitizers in the photovoltaic devices. The starting material for preparing organic dves by standard reactions is carbazole and phenothiazine. The dyes' status changes are assessed by spectrophotometric measurements of the organic photosensitizers in acetonitrile and on a  $TiO_2$  and  $rGO/TiO_2$  substrate. The maximum absorption wavelength for Dyes 1-4 in acetonitrile, TiO<sub>2</sub> films, and rGO/TiO<sub>2</sub> was investigated. Finally, the proposed dyes used as a photosensitizer in a dye solar cell structure in the presence of  $rGO/TiO_2$  and their photovoltaic properties were investigated. Prog. Color Colorants Coat. 15 (2022), 123-131<sup>©</sup> Institute for Color Science and Technology.

## **1. Introduction**

Today, energy and the environment are two dimensions of the international community. Eco-friendly and sustainable energy resources attracted more attention due to clean and endless energy [1, 2]. Photovoltaic technology (PV) is an optimistic and explored area that has exhibited successive year-on-year growth over the last decade. The dye-sensitized solar cell is one of the critical branches of the emerging third-generation photovoltaic technology that has been extended into a most inexpensive energy generator and due to the development of various efficient components [3, 4]. The standard DSSC comprises the dye-sensitized semiconductor photoanode, electrolyte, and counter electrode [5]. The dye-sensitized semiconductor photoanode is used to adsorb dyes, collect and transport photo-induced electrons. It acts as a bridge that conveys electrons from dyes to external circuits and plays a significant role in DSSC [6, 7]. Many studies have focused on developing optimal and new nano-materials for photoanode preparation.

Merazga et al. investigated the effect of reduced graphene oxide (rGO) on the performance of rGO/TiO<sub>2</sub> composite-based dye-sensitized solar cells (DSSCs) is studied. rGO was mixed with TiO<sub>2</sub> in an aqueous solution at different mass proportions, from 0 to 5 %,

to prepare the rGO/TiO<sub>2</sub> composite films. The optical properties of the rGO/TiO<sub>2</sub> films were correlated to the photovoltaic characteristics of associated DSSCs. The optical band-gap Eg of the rGO/TiO<sub>2</sub> film decreases linearly, whereas the efficiency of the DSSC increases linearly with increasing rGO proportion [8]. Patil et al. fabricate a photovoltaic device used organic indoline DN350 and reduced graphene oxide (rGO)-TiO<sub>2</sub>. These precursors (materials) were applied as a sensitizer and photoanode, respectively. Our photovoltaic results revealed that the modified rGO-TiO<sub>2</sub> composite NFs exhibited higher power conversion efficiency (PCE) than the pristine-TiO<sub>2</sub> NFs. Our optimized sample containing 4 mgrGO-TiO<sub>2</sub> NFs showed the best performance with 4.43% PCE, higher than the pristine-TiO<sub>2</sub> NFs (3.83 %) [9]. Ding et al. produced reduced graphene (rGO)-TiO<sub>2</sub> nanocomposite at room temperature. Reduced graphene provides a highway for electron transportation. The conversion efficiency of a 30 % increase (to 7.89 %) is obtained compared to that of the pure  $TiO_2$  photoanode [10].

This paper reports on a study that prepares DSSC from graphene oxide (rGO)-TiO<sub>2</sub> and organic dyes based on indoline. They were employed as photoelectrode and sensitizers, respectively. The DSSC structure and its efficiency have been studied used platinum as a counter electrode and iodide-triiodide as the electrolyte. The spectrophotometric properties were examined in organic dyes included solution and on the photoanode substrate. The electrochemical measurement is also analyzed. The DFT data were used to determine the highest occupied molecular orbital, lowest unoccupied molecular orbital levels, and band gaps of these organic dyes. The structures of the dye molecule are given in Figure 1.



Figure 1: Chemical structure of organic dyes.

#### 2. Experimental

#### 2.1. Materials and instrumentation

All compounds used in this study were of analytical grade unless otherwise stated. The synthesis route and complete characterization of intermediates and organic dyes [11] and nanoparticles have been described previously [12]. UV-Visible spectrophotometry was carried out on Cecil 9200 double beam transmission spectrophotometer. Its result is obtained Molar extinction coefficients and absorption maxima.

# 2.2. Dye-sensitized solar cells (DSSCs) assembly and photovoltaic characteristics

A nanocrystalline film was coated on a transparent glass support. The dye was adsorbed by dipping the coated glass in a  $5 \times 10^{-5}$  M ethanolic solution of the dye for 12 hr. The visible bands in the dyes' absorption spectrum after adsorption on the nanocrystalline film only appeared after the work electrodes were dipped in the dye solution for at least 12 hr. Finally, the film was washed with an acetonitrile-ethanol 1:1 mixed solution. Acetonitrile-ethylene carbonate (v/v=1:4) containing tetrabutylammonium iodide (0.5 mol dm<sup>-3</sup>) was used as the electrolyte. The dye-adsorbed nanocrystalline electrolyte solution were assembled into a sealed sandwich-type solar cell [13, 14].

An action spectrum was measured under monochromatic light with a constant photon number  $(5 \times 10^{15} \text{ photon cm}^2.\text{s}^{-1})$ . J-V characteristics were measured under illumination with AM 1.5 simulated sunlight (100 mW cm $^2$ ) through a shading mast (5.0 mm×4 mm) using a Bunko-Keiki CEP-2000 system.

#### 3. Results and Discussion

Indoline dyes D1-D4 were synthesized, as shown in Figure 1. The aldehydes intermediates were prepared by a Vilsmeier reaction of N-phenylcarbazole and N-phenylphenothiazine with phosphoryl chloride (POCl<sub>3</sub>) in DMF and were allowed to react separately with malonic acid or cyanoacetic acid in the presence of piperidine to give Dye 1 to Dye 4. The final reaction for dye preparation was the condensation of the respective aldehydes with cyanoacetic acid (or malonic acid). It undertook the Knoevenagel reaction in the presence of piperidine. The phenothiazine itself is bent along the N-S axis. As confirmed by X-ray structure

analysis, the nitrogen atom in phenothiazine moiety induces a nonplanar geometry similar to the SP<sup>3</sup> hybridized, pyramidal nitrogen [15].

Conversion efficiency depends mainly on sensitizers, so the recent development in this respect has led to dyes that absorb across the visible spectrum leading to higher efficiencies. The wavelength of maximum absorption  $(\lambda_{max})$  and the molar extinction coefficients ( $\varepsilon_{max}$ ) for the eight dyes in CH<sub>3</sub>CN are listed in Table 1, together with  $\lambda_{max}$  of the corresponding dyes adsorbed on a nanostructure film. The absorption peaks for synthesized dyes can be assigned to an intramolecular charge transfer between the heterocycle-based donor and the acrylic or cynoacrylic acid, providing an efficient chargeseparation in the excited state [5, 16]. Under similar conditions, Dyes 3 and 4 show a bathochromic shift in the absorption peak, which can be attributed to the extra electron-donating effect of the sulfur atom in these dyes. Such bathochromic shifts in the adsorption spectra imply lower energy absorption. For the cyanoacrylic acid based dyes, when an electron acceptor (-CN) was linked to the vinyl bridge, the  $\lambda_{max}$ has a bathochromic shift. This shift of the maximum absorption peak arises from the fact that one more electron acceptor (-CN) enhances the electronwithdrawing ability of electron acceptors and lowers the lower unoccupied molecular orbital (LUMO). Therefore, these changes has reduced the gap between higher unoccupied molecular orbital (HOMO) and LUMO states [17]. Upon dye adsorption onto a  $TiO_2$ and rGO/TiO<sub>2</sub> composite surface, the wavelength of maximum absorption is bathochromically shifted synthesized dyes as compared to the corresponding spectra in solution, implying that dyes adsorbed on to photoanode surface contain partial J-type aggregates [18,19]. The molar extinction coefficients of synthesized dyes in CH<sub>3</sub>CN at their respective  $\lambda_{max}$  are also shown in Table 1, indicating that these novel dyes have good light-harvesting abilities.

Optical absorption spectra of rGO-TiO<sub>2</sub> nanocomposite were carried out to calculate the bandgap energy. When rGO is incorporated with  $TiO_2$ , improved absorption occurs in the region of 200-380 nm, which is due to the  $\pi$ - $\pi$ \* transition. This absorption band has resulted from the electronic transition from O2p antibonding orbital to Ti3d bonding orbital. The band-gap energy of rGO-TiO<sub>2</sub> nanocomposite is determined to be 3.1 eV from the linear fit of the Tauc plot, which confirms the redshift in the absorption spectrum. The absorption shift towards the visible region is due to the incorporation of rGO in TiO<sub>2</sub> nanocomposite, which can be used in photovoltaic applications to harvest visible light photons [20, 21].

Density functional theory (DFT) calculations were performed at the B3LYP/6-31G level to investigate dyes in the mood of ground statesatmolecular levels [7]. Optimized structures, distributions of highest occupied molecular orbitals (HOMO), and lowest unoccupied molecular orbitals (LUMO) are charted with the respective molecular orbital amplitude in Figure 2. According to the theoretical calculation, the HOMO values are calculated of 5.68, 5.96, 5.32 and 5.58 eV for dyes 1, 2, 3, and 4, respectively. They are in the well reflecting the donor moieties' electrondonating strength and the LUMO values of dyes. The LUMO values are registered of 1.86, 2.53, 1.97, and 2.65 eV for dyes 1, 2, 3, and 4 tandemly (Table 2). A slight anomaly of different values of HOMO-LUMO of dyes is related to the part of molecules also covering the cyano group fragment.

Dye	$\lambda_{max} (nm)^a$	$\epsilon_{max} (M^{-1} cm^{-1})$	$\lambda_{max} (nm)^{b}$	$\lambda_{max} (nm)^{c}$
1	386	23898	406	405
2	395	22645	419.5	414
3	404	18746	422	419
4	417	19430	436	428

Table 1: Absorption properties of synthesized dyes.

a) in solution (CH<sub>3</sub>CN); b) on TiO<sub>2</sub> and c) on rGO/TiO<sub>2</sub>

Dye	HOMO, eV	LUMO, eV	Transition	Energy, eV	Main transition (%)
			$S_0 \rightarrow S_1$	3.51	H→L (4.35%); H+1→L (92.47%)
1	5.68	1.86	$S_0 \rightarrow S_2$	3.79	H→L (60.98%); H+1→L+1 (34.32%)
			$S_0 \rightarrow S_3$	4.12	H-2→L (95.04%)
			$S_0 \rightarrow S_1$	3.09	H→L (9.46%); H+1→L (89.02%)
2	5.96	2.53	$S_0 \rightarrow S_2$	3.42	H→L (83.65%); H+1→L+1 (5.51%)
			$S_0 \rightarrow S_3$	4.01	H-4→L (2.40%); H-3→L (90.74)
			$S_0 \rightarrow S_1$	2.94	H→L (2.12%); H+1→L (95.27%)
3	5.32	1.97	$S_0 \rightarrow S_2$	3.72	H→L (9.92%); H+1→L+1 (86.34%)
			$S_0 \rightarrow S_3$	3.89	H→L (22.41%); H+1→L+2 (65.46%)
			$S_0 \rightarrow S_1$	2.58	H→L (2.89%); H+1→L (95.85%)
4	5.58	2.65	$S_0 \rightarrow S_2$	3.54	H→L (86.86%); H+1→L+1 (6.15%)
			$S_0 \rightarrow S_3$	3.86	H→L (5.54%); H+1→L+1 (89.40%)

 Table 2: Values of HOMO-LUMO and calculated transition energies for the first spin allowed, spin forbidden transition and compositions of major electronic transitions of Dyes.



Figure 2: Optimized structure at ground state (S<sub>0</sub>) and distributions of Molecular orbitals HOMO and LUMO of dyes 1-4 calculated at package level of B3LYP/6-31G.

On the other hand, in a comparison of molecular structures, the HOMO-LUMO values of dyes 2 and 4 were in the high range, and it is attributed to the HOMO of carbazol that it is a  $\pi$ -type orbital whereas its LUMO is a  $\pi^*$  orbital localized on cyano group. Hence, the lowest electronically excited state is expected to have intermolecular charge transfer (ICT) character. The absorption bands correspond to the HOMO→LUMO transitions for all dyes are presented in Figure 3. The theoretical transition maximum wavelength values were from 350 to 420 nm, and this range was in good agreement with experimental UV-Vis spectra of dyes (Table 1). According to Table 2, it is noted that the different values of energy levels didn't achieve up to 1 eV, relatively the minor energy splitting between H $\rightarrow$ L and H ± n  $\rightarrow$ L ± n (< 1 eV, n is an integer) with different d orbital character. This event will be significantly caused to enhance the transitions [22, 23].

Dye-sensitized solar cells (DSSCs) were constructed and compared to clarify the relationships between organic dye molecules' sensitizing behavior and rGO/TiO<sub>2</sub> thin films. The DSSCs utilized this dye as a sensitizer for nanocrystalline anatase TiO2 and rGO/TiO<sub>2</sub>. Figure 4 shows a typical photocurrentphotovoltage (J-V) curve for cells-based organic dye. The detailed photovoltaic parameters are also summarized in Table 3. The solar energy to electricity conversion efficiency  $(\eta)$  of the DSSCs is calculated from short circuit current  $(J_{sc})$ , the open-circuit photovoltage (Voc), the fill factor (FF), and the intensity of the incident light (Pin) [13, 24, 25].

Table 3: Photovoltaic performance	e of DSSCs based on	organic dyes.
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DSSCs based on TiO <sub>2</sub> [12]							
Dye	V <sub>OC</sub> (V)	$J_{SC}$ (mA.cm <sup>-2</sup> )	FF	η (%)			
1	0.56	8.29	0.64	2.98			
2	0.63	9.44	0.62	3.69			
3	0.63	10.63	0.64	4.31			
4	0.69	12.67	0.67	5.87			
DSSCs based on rGO/TiO <sub>2</sub>							
	DSS	SCs based on rGO/1	TiO <sub>2</sub>				
Dye	DSS V <sub>oc</sub> (V)	6Cs based on rGO/7 J <sub>SC</sub> (mA.cm <sup>-2</sup> )	FiO2	η (%)			
Dye 1	DSS V <sub>oc</sub> (V) 0.63	SCs based on rGO/T J <sub>SC</sub> (mA.cm <sup>-2</sup> ) 8.31	FF 0.65	η (%) 3.40			
<b>Dye</b> 1 2	DSS V <sub>oc</sub> (V) 0.63 0.69	SCs based on rGO/T J <sub>SC</sub> (mA.cm <sup>-2</sup> ) 8.31 9.60	FiO <sub>2</sub> FF 0.65 0.64	η (%) 3.40 4.23			
Dye 1 2 3	DSS Voc (V) 0.63 0.69 0.68	SCs based on rGO/T J <sub>SC</sub> (mA.cm <sup>-2</sup> ) 8.31 9.60 10.77	FiO2 FF 0.65 0.64 0.64	η (%) 3.40 4.23 4.68			



Figure 3: Theoretically predicted UV-Vis spectra of dyes 1-4 of the singlet transitions (according to the basic set of: TD-DFT B3LYP/6-31G).



Figure 4: Current density-voltage characteristics for a) TiO<sub>2</sub> and b) rGO/TiO<sub>2</sub>.

According to the results shown in Table 3, under the standard global AM 1.5 solar condition, the conversion efficiencies of solar cells containing organic (Dye 1-4) based on TiO<sub>2</sub> are 2.98, 3.69, 4.31, and 5.87 percent, respectively. The efficiencies of solar cells containing organic (Dye 1-4) based on rGO/TiO<sub>2</sub> are 3.40, 4.23, 4.68, and 6.06 percent, respectively. The device's conversion efficiency is probably due to the more vital electron withdrawal ability of the combination of cyanine acceptor groups [26]. The conversion efficiency of solar energy to electricity of the present organic dyes could be improved by extending the organic dyes' conjugated length or by incorporating a thiophene  $\pi$ -bridge [27, 28].

rGO is a carbon allotrope with a 2D honeycomb network of sp<sup>2</sup> hybridized carbon atoms. It has to attract application in DSSC because it possesses high electron mobility ( $2 \times 10^5$  cm<sup>2</sup>.V<sup>-1</sup>.s<sup>-1</sup>), high transmittance (97.7%), and a large surface area (2630 m<sup>2</sup> g<sup>-1</sup>). Incorporation of rGO in TiO<sub>2</sub> can form a heterojunction at its interface and induce a small internal electric field that separates the electron-hole pairs from the charge recombination [29, 30]. It has been reported that rGO-TiO2 nanocomposite (functionalized anatase TiO<sub>2</sub>) reduced graphene oxide) gives better electron mobility  $(53,000 \text{ cm}^2.\text{V}^{-1}.\text{s}^{-1})$  and enormous surface area  $(200 \text{ m}^2 \text{ g}^{-1})$ . This leads to the improved electrontransport property and dye adsorption, respectively. However, incorporating a high volume fraction of rGO is not a preferred choice for photoanode application due to its reduced dye anchoring. Because the high withdrawing ability of -COOH moiety of dye sensitizers can anchor efficiently on TiO2 surface rather than rGO sheets [31]. Junling Song et al. reported that, the introduction of the rGO layer between TiO<sub>2</sub> and dye could boost the electron transportation by the formation of rGO-TiO<sub>2</sub> Schottky junction, which enhances the power conversion efficiency up to 6.06 % compared with pristine TiO<sub>2</sub> based device (5.09 %) [32].

According to the above results, graphene has caused significant improvement in our fabricated Schottky barrier diode's various parameters. Graphene has an electron mobility of 10<sup>4</sup> cm<sup>2</sup>.V<sup>-1</sup>.s<sup>-1</sup> at room temperature. So, it is expected to enhance the electron transfer and EHP separation of the composite. This impact of graphene is verified, and get a better insight into the carrier transport mechanism, the I-V curves were further analyzed by evaluating mobility, lifetime, and diffusion length of the carriers. Standard SCLC theory was employed for this purpose. Charge carriers in the graphene composite-based diode had reasonably greater diffusion length than those in the other device, more than 30 % under photo condition. As a result, more charge carriers were generated upon light soaking, thus increasing the photocurrent significantly. In our graphene-TiO<sub>2</sub> composite, graphene acts more like many extended current collectors penetrating the  $TiO_2$  matrix. Electrons can travel a longer distance and finally get collected by an external circuit [33, 34].

#### 4. Conclusions

Four organic dyes with D- $\pi$ -A structure (phenyl as donor and acrylic and cyanoacrylic acid as acceptor group) were assembled as photosensitizers in dyesensitized solar cell devices. rGO/TiO2 was selected as nanostructure for the preparation of photoanode of dyecells. sensitized solar The organic dyes' spectrophotometric properties included solvent, and TiO<sub>2</sub> and rGO/TiO<sub>2</sub> films were investigated. The absorption maxima of synthesized dyes applied on the  $rGO/TiO_2$  film's surface gave a bathochromic effect compared to the corresponding dye spectra in solution. The wavelength of maximum absorption for Dyes 1-4 in acetonitrile are 386, 395, 404, and 417 nm and, on TiO<sub>2</sub> films are 406, 419.5, 422, and 436 nm, and, on rGO/TiO<sub>2</sub> 405, 414, 419, and 428 nm, respectively. Finally, the organic dyes were utilized in constructed DSSCs, and their photovoltaic behaviors were assessed. Solar energy to the electricity conversion efficiency of 2.98, 3.69, 4.31, and 5.87 percent was achieved for synthesized dye 1-4 in DSSCs based on TiO<sub>2</sub>, respectively. Solar energy to the electricity conversion efficiency of 3.40, 4.23, 4.68, and 6.06 percent was achieved for synthesized dye 1-4 in DSSCs based on rGO/TiO<sub>2</sub>. The results show that the overall conversion efficiencies of DSSCs sensitized by all dyes in the presence of rGO/TiO<sub>2</sub> are higher than DSSCs based on TiO<sub>2</sub> nanoparticles.

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