

EFFECT OF π -ELECTRON SYSTEM IN ORGANIC DYES FOR DYE-SENSITIZED SOLAR CELLS

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Abstract: We have evaluated the effect of π -electron system in donor- π -acceptor (D- π -A) based organic dyes and observed the performances in dye sensitized solar cell (DSSC). Various π -conjugation structures with different molecular arrangements with and without possessed π -conjugation system were successfully evaluated with custom molecular organic dyes D-1, U01, AK-01, TA-St-CA, Ba-03 and YF02. The introduction of conjugated π -system in the D- π -A system elevated wide range of absorption spectra from 300 nm to 850 nm wavelength region. The DSSC based on dyes D-1, U01, AK-01, TA-St-CA, Ba-03 and YF02 showed onset absorption of 525 nm, 600 nm, 650 nm, 550 nm, 810 nm and 550 nm correspondingly on the TiO₂ film. A dip at the IPCE spectra in the 350 nm–470 nm wavelength region for dye D-1 and U01 were attributed to the competitive absorption of solar spectrum of I³⁻ solution. The AK-01 and BA-03 dyes showed IPCE of 70% at the 360 nm–620 nm region and subsequently reaches up to a maximum of 84% at 450 nm. The U01 dye depicts IPCE of 70% at 400 nm–560 nm and peaks of 86% at 500 nm within the visible region. The sensitized YF02 dye reaches up to 75% at 350 nm–550 nm. The greater IPCE value of AK-01, TA-St-CA, YF02 and BA-03 in the UV zone suggested that these organic dyes successfully surpassed the dip of IPCE spectrum in that region. The photovoltaic performances of these sensitizers were verified by incident-photon-to-current conversion efficiency (IPCE), open-circuit photovoltage (V_{oc}), short-circuit photocurrent density (J_{sc}) and total Power conversion Efficiency (η). Dyes D-1 and U01 were without any π -linker showed η of 3.34% and 6.01% exhibiting J_{sc} of 6.42 mA cm⁻² and 10.70 mA cm⁻² respectively. Dyes with a π -conjugated oligo-phenylenevinylene unit linker showed η of 6.2% (AK-01), 5.41% (TA-St-CA) and 5.29% (YF02), dye with a fused dithienothiophene π -conjugated linker showed η of 6.38% (Ba-03) respectively. The corresponding J_{sc} of dyes AK-01, TA-St-CA, Ba-03 and YF02 were 15.40 mA cm⁻², 10.10 mA cm⁻², 16.17 mA cm⁻² and 9.19 mA cm⁻² followed by V_{oc} of 0.640 V, 0.718 V, 0.595 V and 0.799 V respectively. The comparative analysis of IPCE reveals the effect of π -electron of these dyes to increase their corresponding light harvesting ability, reduced dye aggregation and low carrier recombination in DSSCs operation.

Keywords: Dye sensitized solar cells, organic dye, incident photon to current conversion efficiency, molecular planarity, π -conjugation, HOMO-LUMO (Highest Occupied Molecular Orbital-Lowest Unoccupied Molecular Orbital).

Introduction

The Sun's energy has been identified as the cleanest energy sources due to its unlimited power supply. Many efforts have been engaged in the search of cost effective and gaining higher conversion efficiency by different device structured solar cells (Chowdhury *et al.*, 2016; Beard *et al.*, 2014). A major revolution

in 1991 with the introduction of 7% efficient dye sensitized solar cells (DSSC) (O'Regan & Grätzel, 1991) which imitates the natural photosynthesis process of production and store energy created a whole in new dimension in photovoltaic technology. In their introduced method, a chromophore (e.g. a dye) is used to sensitize the semiconductor as chlorophyll does in photosynthesis process. DSSC became the new

center of systematic and commercial interest in lieu of silicon photovoltaics for offering more worthwhile phenomenon (Reddy *et al.*, 2014; Ragoussi & Torres, 2015).

Generally, a DSSC comprises of a nanocrystalline photoanode such as TiO_2 (Hagfeldt & Grätzel, 1995; SnO₂ (Ramasamy & Lee, 2010) or ZnO (Rensmo *et al.*, 1997), a platinum electrode (cathode), sensitizer and an electrolyte solution with a dissolved (I^3/I^- ion) redox couple. The sensitizing dye in a DSSC is anchored to a wide-band gap semiconductor. Upon absorbing light (photon) with sufficient energy by the dye, the photoexcited electron rapidly transfers to the conduction band of the semiconductor, which carries the electron to one of the electrodes (Ardo & Meyer, 2009). The redox couple, usually comprised of iodide/triiodide (I^-/I^3^-), then reduces the oxidized dye back to its neutral state and transports the positive charge to the platinized counter-electrode (Hagfeldt *et al.*, 2010).

Therefore, the role of the sensitizer is one of the crucial factors which directly affect the PCE of a DSSC. One of the most intensive studied, molecular structure of organic dyes contains donor- π linker-acceptor (D- π -A) (shown in the Figure 1) pattern which is very suitable in n-type DSSCs, where the acceptor moiety acts as the anchoring group (Casanova, 2011).

In general, for improving DSSCs performances, the organic dyes must show high short-circuit photocurrent (J_{sc}), high open-circuit photovoltage (V_{oc}) and high fill factor

(FF). During the evaluation of the DSSCs performances, it was observed that the dyes with minimal driving force of 0.2 eV with E_{0-0} energy gap of 1.4 eV (minimum ground-state oxidation potential ($S^{+/0}$) of -5.4 eV, maximum excited-state oxidation potential ($S^{+/*}$) of -4.0 eV involved in efficient electron injection and regeneration process into DSSC operations (Islam *et al.*, 2003). The E_{0-0} energy gap of the black dye is 1.6 eV which was reported with the highest PCE of 11.1% (Han *et al.*, 2012; Ogura *et al.*, 2009; Chen *et al.*, 2005; Kuang *et al.*, 2007). However, till date the successful organic dyes contained energy gap of $E_{0-0} > 2.0$ eV. Another major drawback of organic dye is highlighted with the formation of π to π aggregation on TiO_2 surface which reduces the electron injection rate into the TiO_2 electrode, and eventually leads to low V_{oc} . Although several attempts have been taken to improve the V_{oc} with addition of additives such as DCA (Deoxycholic acid), DMPII (1, 2-dimethyl-3-propylimidazonium) and TBP (4-tertbutylpyridine) with the electrolyte but the J_{sc} of the DSSCs decreased simultaneously. The J_{sc} is correlated with the absorption ability and electron injection efficiency of dyes, while the V_{oc} is considered to be more complicated than J_{sc} . The charge injection and recombination process are closely correlated with V_{oc} , which means that V_{oc} becomes one of the key factors to enhance the performance of DSSCs. The dye aggregation can also lead to self-quenching and reduce the electron injection rate resulting in a low V_{oc} which can be controlled by reducing dye aggregation on the TiO_2 surface and also

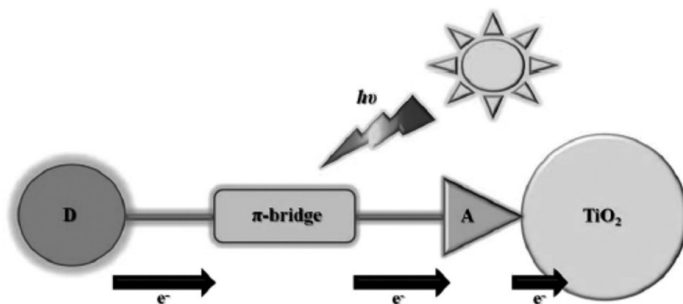


Figure 1: Schematic diagram of donor- π -acceptor organic sensitizers

by increasing the molecular planarity between donor and π -conjugation system. For an example, a novel series of D- π -A sensitizers were K-1, K-2 and their carbazole analogue MK-3 (Zhao *et al.*, 2014). DSSCs based on K-dyes showed sufficiently good efficiency of 6.62% and 6.73% than MK-3 (4.7-4.8%) without significant decrease of V_{oc} (0.70-0.71 V) even though K-dyes had greater band gap than MK-3. This outcome can be attributed to the different π -conjugation system in those dyes framework producing varieties rotation angle due to contain different atoms in the π -system itself. Therefore, the π -system has been considered as vital factor in conversion process since it plays an essential role in tuning the main properties of the sensitizer. The challenge is to understand how and why different chemical modifications in the conjugated linker tune the electronic structure of the chromophores should guide experimental studies to keep improving the IPCE of organic dyes.

In this study, we report on the comparative performance of organic dyes based on with or without different π -conjugation system. We have selected indoline based dyes D-1 and U01 (without π -linker), with a π -conjugated oligophenylenevinylene unit linker AK-01, TA-St-CA, YF02 and with a fused dithienothiophene π -conjugated linker dye Ba-03 as shown in the Figure2 to investigate effect of different π -spacer in DSSCs performance (Akhtaruzzaman

et al., 2011; Akhtaruzzaman *et al.*, 2013; Akhtaruzzaman *et al.*, 2013; Yang *et al.*, 2012).

Methodology

The synthesized and device fabrication process of dyes D-1, U01, AK-01, TA-St-CA, YF02 and Ba-03 were reported elsewhere (Akhtaruzzaman *et al.*, 2011; Akhtaruzzaman *et al.*, 2013; Akhtaruzzaman *et al.*, 2013; Yang *et al.*, 2012). The optical properties of DSSC were recorded by using the Beer-Lambert Law (Swinehart, 1962) with 1 cm path length quartz glass via the solvent of ethanol. The photoanode was prepared according to published procedures (Islam *et al.*, 2015). The conducting glass substrates of fluorine-doped tin oxide (FTO) (resistance of 8–10 Ω^{-2}) with optical transmission of higher than 80% in the visible range were used from commercial sources. The double layer nanocrystalline TiO₂ film was prepared through a screen printing method with a specification of (15+5) μm in thickness (0.25 cm² cell area), 8- μm transparent layer with particles size of \approx 20 nm and a 5- μm scattering layer of TiO₂ particles (\approx 400 nm). After that, the films were sintered at 500°C for 1 h. Before fabricating the solar cell, it was further treated with 0.1M solution of HCl. The thicknesses of films were measured with a Surfcom 1400A surface profiler (Tokyo Seimitsu Co. Ltd.). The 0.2 mM solution of organic dye was prepared in the mixture solvent of acetonitrile/ tert-butyl alcohol in 1:1 (v/v)

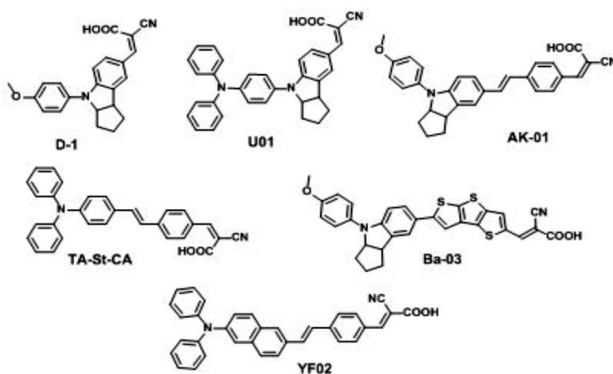


Figure 2: Molecular structure of organic dyes D-1, U01, AK-01, TA-St-CA, Ba-03 and YF0

Table 1: Molar extinction co-efficient of dyes D-1, U01, AK-01, TA-St-CA, YF02 and Ba-03 at their corresponding maximum absorption in ethanol solution

Dye	Maximum Absorption Peak, λ (nm)	Molar Extinction Coefficient, ϵ ($M^{-1}cm^{-1}$)	E_{0-0} (eV)
D-1	440	2.66×10^4	2.33
U01	450	3.4×10^4	2.06
AK-01	436	3.4×10^4	2.39
TA-St-CA	412	3.1×10^4	2.25
BA-03	485	4.0×10^4	1.86
YF02	300	3.2×10^4	2.25

ratio. Then, the TiO_2 film was dipped into the solution at $25^\circ C$ for 48 h. The Platinum-coated electrode and photoanode were separated using 40 μm thick of Surlyn polymer by heating process. An electrolyte containing 0.6M dimethylpropyl-imidazolium iodide, 0.05M I_2 , 0.1M LiI and 0.4M tert-butylpyridine in acetonitrile was used in all the DSSCs. The current-voltage characteristics were measured by using a black metal mask with an area of 0.25 cm^2 under AM 1.5 sunlight (100 Wcm^{-2} , WXS-155S-10: Wacom Denso Co. Japan). The IPCE spectra were measured with a monochromatic incident light of 1×10^{16} photons cm^{-2} in direct current mode (CEP-2000BX, Bunko-Keiki).

Results and Discussion

Optical Properties

Molar extinction co-efficient of the custom molecular organic dyes are presented in Table 1. Except the dye D-1, all dyes showed higher molar extinction co-efficient values with maximum absorption maxima up to 650 nm as shown in Figure 3. With introducing the π -conjugation, the absorption maxima red-shifted except dye D-1 and U01. The E_{0-0} energy gap of AK-01 was lower and showed red-shifted absorption maxima with higher molar absorption coefficient, compared with TA-St-CA (as shown in Table 1 and Figure 3). This is due to stronger donating ability of indoline moiety as well. The absorption band edge of Ba-03 was extended up

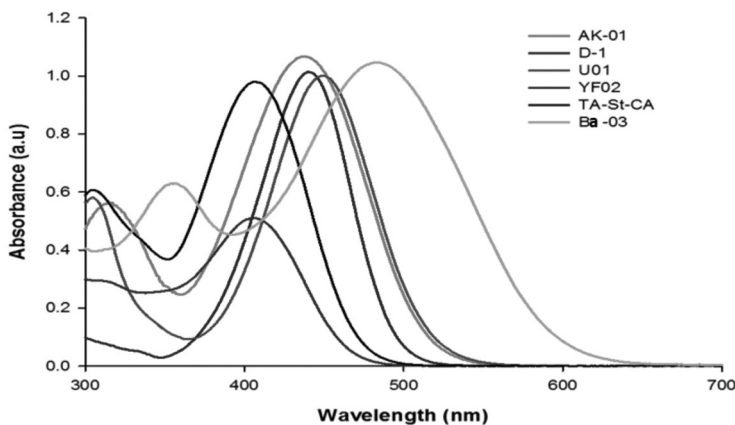


Figure 3: UV-Visible absorption spectra of D-1, U01, Ak-01, TA-St-CA, YF02 and Ba-03 in ethanol

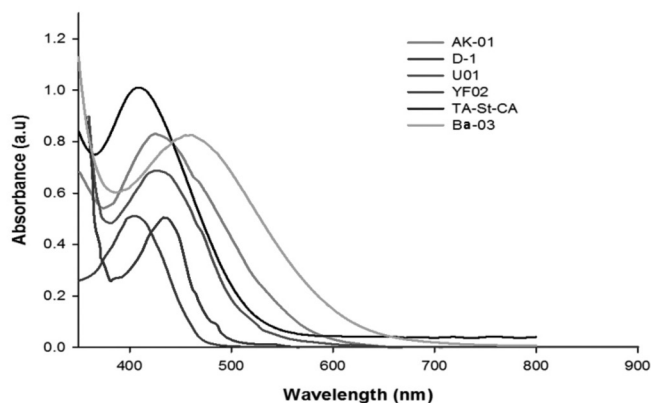


Figure 4: Absorption spectra of dyes D-1, U01, AK-01, TA-St-CA, YF02 and Ba-03

to 650 nm wavelength region. The absorption maxima (λ_{max}) of U01 showed a superior light-harvesting efficiency only compared to the reference dye D-1. The absorption band edge of U01 was stretched up to 565 nm and red shifted by 0.17 eV. The wavelength maxima of YF02 were 406 nm in ethanol and 550 nm onset in TiO_2 .

As shown in Figure 4, the absorption spectra of sensitizers onto the TiO_2 films. The similar red-shifted absorption maxima was also observed on TiO_2 surface of dyes AK-01 and TA-St-CA although the absorption peak slightly blue-shifted (shown in Figure 2) compared to the solution state. The absorption maxima of U01 showed a superior light-harvesting efficiency compared to D-1. Low-lying absorption peaks, corresponding to the π - π^* transition were exhibited at 450 nm for U01 with a higher molar extinction coefficient of dye D-1 at 440 nm. U01 showed broad absorption spectrum similar to that in solution. However, the absorption maxima were slightly blue-shifted due to the interaction between the carboxylate group and TiO_2 . Dyes Ba-03 and YF02 also showed significant red shifted absorption extended up to up to 700 nm and 550 nm respectively. All these dyes were designed with same anchoring group such as cyanoacrylic and carboxylic acid but different π -conjugation system with different donor moiety. The blue shift absorption maxima of all dyes were due to the strong binding on

TiO_2 nanoparticles through the $-\text{COOH}$ group. In general, wider absorption maxima, higher molar extinction coefficient and appropriate matching of the HOMO-LUMO (Griffith & Orgel, 1957) energy level with tri-iodide and TiO_2 are favorable for improving the DSSCs performances (Hara *et al.*, 2003).

Photovoltaic Performances of Dyes in DSSCs

The monochromatic IPCE spectra were shown in the Figure 5 with corresponding wavelength for DSSCs based using individual dye AK-01, D-1, U01, TA-St-CA, YF02 and Ba-03 under AM 1.5 irradiation (100 mW cm^{-2}). All dyes showed absorption ranging within 300 nm–850 nm regions. The dip in the IPCE spectra at 350 nm–470 nm wavelength regions of dye D-1 and U01 were attributed to the competitive absorption of solar spectrum of I_3^- solution. This dip at the ultraviolet zone greatly limited the performance of DSSCs which might be attributed to the absence of π -conjugation system compare to other dyes. The dye AK-01 and BA-03 showed IPCE of 70% in 360 nm–620 nm region, reaching up to a maximum of 84% at 450 nm. The U01 dye showed IPCE of 70% over the visible region of 400 nm–560 nm and peaks of 86% at 500 nm. The sensitized YF02 dye's IPCE reached up to 75% at 350 nm–550 nm. The greater IPCE value of AK-01, TA-St-CA, YF02 and Ba-03 in the UV zone suggested that these organic dyes successfully surpassed the dip of IPCE spectrum

in their region. During our systematic studies on organic dyes, we found that the reported dye TA-St-CA showed a η of 5.41% with band gap of 2.25 eV (Akhtaruzzaman *et al.*, 2011) whereas the authors claimed to achieve the efficiency of 9.1% possessing band gap of 2.4 eV (Hwang *et al.*, 2007).

The photo-current characteristics of DSSCs using AK-01, D-1, U01, YF02, TA-St-CA and Ba-03 were carried out under at AM1.5 sunlight illumination (100 mW cm^{-2}) as shown in Figure 6. The J_{sc} , V_{oc} , FF and η for DSSCs are summarized in Table 2. The widest absorbing Ba-03 showed highest efficiency of 6.38%, which was slightly higher than that of AK-01. Although the J_{sc} of dyes Ba-03 was identified almost to be similar to

AK-01, but the V_{oc} further decreased compared to AK-01 as shown in I - V spectrum in Figure 6. However, the IPCE of Ba-03 covered wider wavelength region (400 nm-600 nm) with the quantum efficiency of around 85% as shown in Figure 6. The IPCE and I - V spectra of YF02 dye clearly indicates the structural effects due to the attached oligo-phenylenevinylene π -linker on DSSCs performances.

It was also observed that the planar π -conjugated unit (Ba-03) showed higher efficiency compared to twisted π -conjugated unit (YF02) and without π -conjugated unit (D-1 and U01) as shown in Table 2. However, the I - V graphs (as shown in Figure 6) showed that the diphenyl containing dye YF02 achieved higher V_{oc} (0.799V) compared to indoline or triphenyl

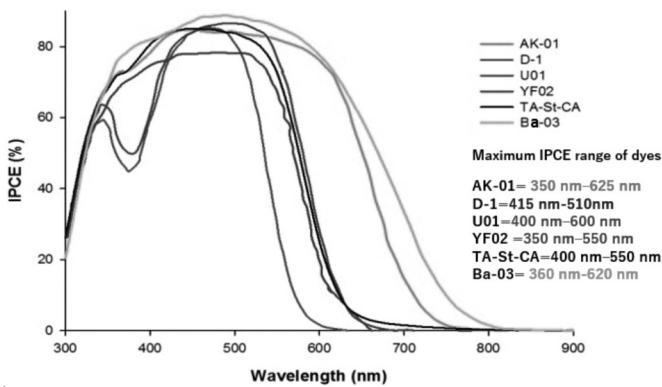


Figure 5: IPCE of DSSCs based on D-1, U01, AK-01, TA-St-CA, YF02 and Ba-03

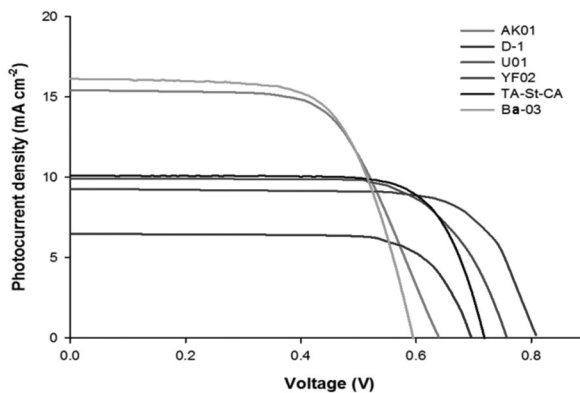


Figure 6: I - V spectra of DSSC based on D-1, U01, AK-01, YF02, TA-St-CA, Ba-03

Table 2: Current-Voltage performance of DSSCs sensitized with individual dye

Dye	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF	η (%)
D-1	6.42	0.705	0.750	3.34
U01	10.70	0.758	0.740	6.01
AK-01	15.40	0.640	0.630	6.20
TA-St-CA	10.10	0.718	0.746	5.41
BA-03	16.17	0.595	0.663	6.38
YF02	9.19	0.799	0.721	5.29

containing donor which depicted remarkable effects of molecular planarity between donor and π -linker restricting recombination. The dye with higher planar π -conjugation structures confirms stronger π - π interactions compared to non-planar structure. The strong π - π interactions of organic sensitizers on TiO₂ surface may cause self-quenching to decrease the DSSCs performances. Although low planar π -conjugated molecule increases the V_{oc} in DSSCs through the blocking of I³⁻ ions towards the TiO₂ surface but simultaneously the efficient intramolecular charge decreases, leading to low J_{sc} .

Conclusion

The performances of organic dye based DSSCs were evaluated in this work. We have presented some useful comparative analysis to obtain highly efficient sensitizers by independently introducing π -linker. The twisted indoline structure between donor and linker of AK-01 enhanced light harvesting ability, reduced dye aggregation with increasing efficiency of 6.2%. YF02 showed higher V_{oc} due to their twisted geometry but reduces the J_{sc} . The sensitizers U01 were low in molecular weight and yielded an efficiency of 6.01%. The research on analogous fused dithienothiophene based dye suggested lower performances due to their loss in charge transfer and low electron injection from excited dye to the corresponding layer. The efficient charge transport of the custom synthesized dyes has provided insights into designing and

introducing precautions steps for newer organic molecules to be used in DSSCs.

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