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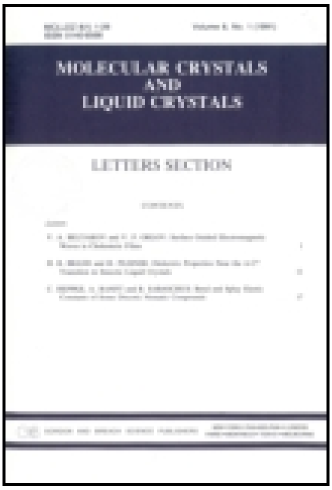

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Performance of Solution Processed Organic Photovoltaic Cells Using A-D-A Type Small Molecular Donors

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*The performance of solution processed bulk heterojunction (BHJ) organic photovoltaic cells (OPVs) has been remarkably improved during the past two decades. Most of BHJ OPVs were produced using polymeric donor materials, since the polymeric donor materials can provide the advantage of easily tailoring the morphology of photoactive films of BHJ OPVs. However, the difficulty in synthetic reproducibility and purification of polymeric donor was main drawback to overcome. In this study, we designed and synthesized A-D-A type small molecular donor materials (BDT(IId)$_2$) containing alternatively linked benzodithiophene (BDT) and Isoindigo units. Also, we investigated the effects of the substituent and alkyl chains of on the efficiency of OPVs . Finally, 2.46% of power conversion efficiency (*PCE*) can be obtained using a mixture of synthesized small molecular donor and PCBM as an active layer with a* Voc *of 0.99 V, a* Jsc *of 6.82 mA/cm^2, and a* fill factor *of 0.36.*

Keywords Organic photovoltaic cells; OPVs; small molecular donor; BDT(IId)2; A-D-A (Acceptor-Donor-Acceptor)

1. Introduction

Organic photovoltaic devices (OPVs) have potential of effective renewable energy sources, and therefore have been intensively studied in academic and industrial [1–3]. Because solution process have low manufacturing cost and high throughput by roll-to-roll processibility [4, 5]. Most of researchers focused on the development of bulk heterojunction (BHJ) based solution processed OPVs using a mixture of conjugated polymer and PCBM [6, 7]. Recently, 8% *PCE* (Power Conversion Efficiency) of OPVs were reported at BHJ devices [8, 9]. To enhance power conversion efficiency, many study used wide band gap polymers

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to absorb long wavelength light for making many excitons [10, 11]. And polymeric donors were well known to form an efficient BHJ by phase separation. Thus, BHJ devices with polymeric donor showed good charge separation and collection [12, 13]. Because of benefit using polymer donor in BHJ devices, BHJ OPVs using small molecular donor have received low attention [14, 15]. However, the difficulty in synthetic reproducibility and purification of polymeric donor was main drawback to overcome.

Although power conversion efficiency up to 8% has been achieved recently with small molecular donors such as DR_3TBDT_2T [16], a major issue for the study of photovoltaic materials is their low light harvesting at short wavelength region due to the narrow absorption of the active layer. In order to increase the efficiency of light harvesting from 300 to 400 nm, the active layer should have higher light absorption region especially around 300–400 nm for high absorption coefficients and the further improved efficiency of organic photovoltaic cells. Incorporating acceptor moieties into molecular conjugated backbones to form donor–acceptor systems is one of the most effective approaches [17].

In this study, we designed and synthesized A-D-A type small molecular donor materials for OPVs, which have Isoindigo (IId) units at both side of benzodithiophene (BDT). BDT and IId unit are well known electron donating and accepting materials, respectively [18, 19]. By incorporation of IId with BDT, band gap can be controlled to wide region and absorption in short wavelength from 300 to 400 nm can be improved. Also, we studied on the photovoltaic effect of alkyl substituent attached in BDT and IId.

Herein, we reported new isoindigo containing small molecules (SM), $BDT(IId)_2$ based small molecular donor materials and their the optical and electrochemical properties with variation of alkyl substituent by UV/Visible spectrophotometer and cyclic voltamometer (CV), respectively. Finally, we fabricated OPVs using one of synthesized A-D-A type $BDT(IId)_2$ based small molecules as an electron donor materials and compared photovoltaic performance of each devices.

2. Experimental Details

2.1. Materials

(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl)bis(trimethylstannane) and (4,8-bis(5-(1-octyloxy)benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl)bis (trimethylstannane) were purchased from Solarmer Materials. 2-Ethylhexyl bromide, 1-octylbromide, Pd_2dba_3, $P(o\text{-}tol)_3$, toluene, and 1,2-dichlorobenzene were purchased from Aldrich. Fullerene (C60) was purchased from BuckyUSA. PEDOT-PSS (AI4083) was purchased from Heraeus. All chemicals were used without further purification. (E)-6-bromo-1, 1′-bis(2-ethylhex-1-yl)-[3,3′-biindolinylidene]-2,2′-dione and (E)-6-bromo-1,1′-bis(1-ocxyl)-[3,3′-biindolinylidene]-2,2′-dione were synthesized in similar ways according to the methods described in previous reports[20].

2.2. Measurements

^1H NMR spectra were recorded using Bruker 300 spectrometer. The UV/Vis absorption spectrum of each compound was measured in solution state dissolved in chloroform by SHIMADZU UV-2550. Electrochemical properties of compounds were measured by Cyclic Voltametry (CV, WonATech BAS 100B). The CV was performed in a 0.1 M chloroform solution containing electrolyte of $Bu_4N(PF_6)$. A molded platinum electrode (0.3 mm diameter) was used as a working electrode, a Pt wire was used as a counter electrode, and a Ag/Ag+ was used as the reference electrode. And solution of reference electrode was used with 0.01 M silver nitrate ($AgNO_3$). CV was measured positive state at room temperature. Measured curve was shown for highest occupied molecular orbital (HOMO) energy level. And we predicted lowest unoccupied molecular orbital (LUMO) energy level using optical band-gap. Atomic force microscopy (AFM) images were taken with a Digital Instruments Nanoscope IV operating in tapping mode in air. BHJ film morphology of a mixture of one of compound 1–4 and PCBM is measured AFM (Digital Instruments). The BHJ film formed on indium tin oxide (ITO) glass. First, poly(3, 4-ethylenedioxythiophene)poly(styrenesulfonate) (PEDOT:PSS) (Clevious AI 4083) was spin-coated on ITO galss and annealed at 150°C for 18 min. Finally, a mixture of SM donor and PCBM was spin-coated on the prepared PEDOT:PSS film and then annealed at 90°C for 10 min.

2.3. Synthesis of BDT(IId)$_2$ (Compound 1-4)

2.3.1. General Procedure of Compound 1-4. In a 100ml dried flask, (E)-6-bromo-1,1′-bis(2-ethylhex-1-yl)-[3,3′-biindolinylidene]-2,2′-dione or (E)-6-bromo-1,1′-bis(1-ocxyl)-[3,3′-biindolinylidene]-2,2′-dione (0.52 mmol), (4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl)bis(trimethylstannane) or (4,8-bis(5-(1-octyloxy)benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl)bis(trimethylstannane) (0.26 mmol), Pd_2dba_3 (5 mg, 2 mmol%), $P(o\text{-}tol)_3$ (7 mg, 8 mmol%) were dissolved in 40 mL of anhydrous toluene. The reaction mixture was heated at 100°C and stirred for 24 h. Then it was poured into water and extracted with ethylacetate. Removal of the solvent and column purification on silica gel using dichloromethane/hexane (2/5) offered the compound **1–4**.

***Compound 1** (yield: 24 %)*

^{1}H-NMR 300 MHz ($CDCl_3$) δ (ppm) 9.15-9.06 (m, 4H), 7.80 (d, J = 3.6 Hz, 2H), 7.42 (d, J = 3.6 Hz, 2H), 7.20-7.14 (m, 4H), 7.00-6.93 (m, 4H), 6.85 (s, 2H), 6.70-6.61 (m, 2H), 3.70-3.64 (m, 8H), 3.05-2.91 (m, 4H), 1.82-0.90 (m, 90H)

***Compound 2** (yield: 25%)*

^{1}H-NMR 300 MHz ($CDCl_3$) δ (ppm) 9.12-9.08 (m, 4H), 7.81 (d, J = 3.3 Hz, 2H), 7.43 (d, J = 3.3 Hz, 2H), 7.20-7.16 (m, 4H), 7.03-6.96 (m, 4H), 6.85 (2, 2H), 6.66 (d, J = 7.8 Hz, 2H), 3.87-3.67 (m, 8H), 3.00 (d, J = 6.3 Hz, 4H), 1.83-0.89 (m, 90 H)

***Compound 3** (yield: 66%)*

^{1}H-NMR 300 MHz ($CDCl_3$) δ (ppm)9.17 (q, J = 11.1 Hz, 4H), 7.71 (s, 2H), 7.38-7.29 (m, 4H), 7.06-7.01 (m, 4H), 6.75 (d, J = 7.8 Hz, 2H), 4.24 (d, J = 5.1 Hz, 4H), 3.77 (d, J = 7.2 Hz, 4H), 3.71-3.66 (m, 4H), 1.91-0.90 (m, 90 H)

***Compound 4** (yield: 76%)*

^{1}H-NMR 300 MHz ($CDCl_3$) δ (ppm) 9.19 (d, J = 8.4 Hz, 4H), 7.66 (s, 2H), 7.30 (d, J = 8.4 Hz, 4H), 7.04-6.97 (m, 4H), 6.73 (d, J = 7.8 Hz, 2H), 4.23 (d, J = 5.1 Hz, 4H), 3.86 (t, J = 7.2 Hz, 4H), 3.77 (t, J = 7.2 Hz, 4H), 1.90-0.86 (m, 90 H)

2.4. Device Fabrication

The SM photovoltaic devices were formed in sandwiched structure of BHJ between a transparent anode and cathode. Transparent anode used ITO having 10 Ω/sq of sheet resistance. Patterned ITO glasses were prepared for sonication process with detergent in distilled water and boiling process with acetone and isopropanol. After this process, ITO glasses were cured with UV/Ozon during 20 min. On the prepared ITO glasses spin coated by PEDOT:PSS at 40 nm followed thermal treatment at 150°C for 18 min. Active layers were spin coated by mixture of SM donor and PCBM with chloroform followed thermal

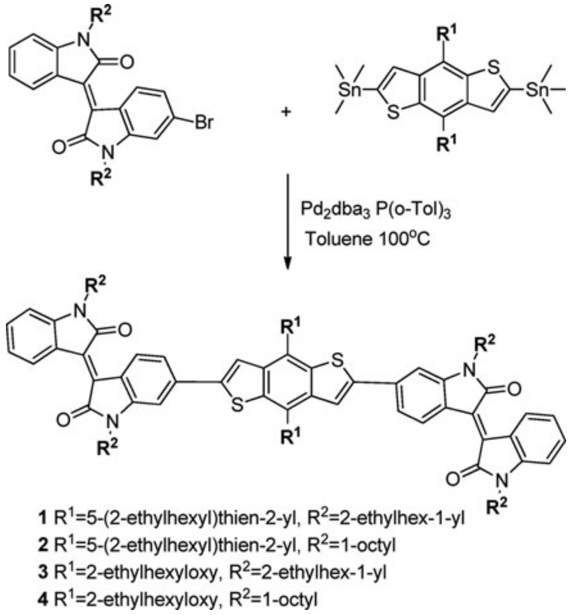


Figure 1. Structure of BDT(IId)$_2$s.

treatment at 90°C for 10 min. Finally, metal cathode was deposed by thermal evaporation consisted with LiF and Al approximately 0.5 Å and 120 nm thick.

2.5. *J-V Measurements*

Photocurrent-voltage curve measurements of each device were performed at room temperature. The currentvoltage *I-V* curves of the devices were obtained using a Keithley 2400 Source Measure Unit under AM 1.5 simulated light (ORIEL 300W Xenon lamp). The intensity of light (100 mA/cm^2) from the illumination source was confirmed by using a standard silicon photodiode (BS520, Bunkoh-Keiki).

3. Results and Discussion

3.1. *Design and Synthesis*

For enhancement of light absorption and mobility of donor materials, we designed A-D-A type donor by using electron donating part with BDT and electron withdrawing part with IId. And we synthesized four small molecular donor compounds (1–4) based on BDT(IId)$_2$ with variation of alkyl substituent of each moiety to compare their photovoltaic performance. Their structure was consists of one BDT at center part and IId at each side part. BDT has two different kinds of substituent, one is 2-(2-ethylhexyl)thiophene and the other is 1-octyloxy. Also, IId has different alkyl chains with 2-ethylhexyl and 1-octyl groups (Fig. 1).

Compound **1–4** were prepared by Stille reaction of bis-trimethylstannanyl BDT derivative and *mono*-bromo isoindigo derivative in the presence of Pd$_2$dba$_3$/P(o-Tol)$_3$ catlayst with 24–76% yields. The chemical structure of synthesized compound **1–4** was characterized by ^{1}H NMR spectroscopy.

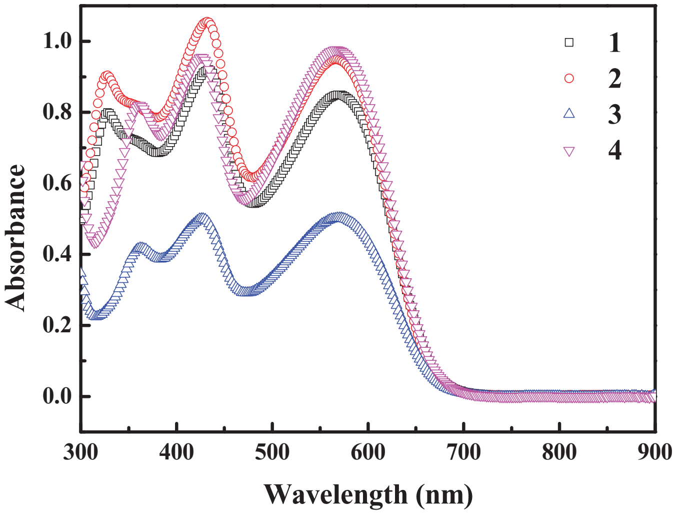


Figure 2. UV absorption spectra of BDT(IId)$_2$s (compound 1–4) in chloroform at 10^{-5} M.

3.2. Photophysical and Electrochemical Properties

Figure 2 and Table 1 show the UV/Vis absorption spectra of SM donors in chloroform solution. All of synthesized A-D-A type donors (compound **1–4**) show similar on-set of UV/Vis absorption at 660 nm. As a result, we confirmed that each compound showed same optical bandgap, which are about 1.86 eV. As a predict, isoindigo group can be used as efficient electron accepting part to tune the bandgap in A-D-A type SM donor. 5-(2-Ethylhexyl)thien-2-yl substituted BDT containing compounds, **1** and **2** showed several absorption bands (326, 432, and 570 nm) at short wavelength. And alkyloxy substituents of BDT containing compounds **3** and **4** showed three absorption bands (361, 426, and 567 nm). In addition, linear alkyl substituted IId containing compounds (**2** and **4**) showed much higher absorption property compare to branched alkyl substituted compounds (**1** and **3**). Especially, 2-ethylhexyl-1-oxy substituted compound (**3**) showed relatively low absorption coefficient compared with others, which is resulted from the crystallization of octyl chain

We measured cyclic voltammograms at positive state for −50 mV to 1200 mV for measuring HOMO energy level. Figure 3 and Table 1 show cyclic voltammograms of compound **1–4** in details. HOMO energy levels of compound **1** and **2** were measured by −5.50 and −5.48 eV, respectively. And HOMO energy levels of compound **3** and **4** were measured by −5.38 and −5.37 eV, respectively. We can confirm BDT having same substituent has

Table 1. Optical and electrochemical properties of donor materials

Compound	HOMO (eV)	LUMO (eV)	Abs. on set (nm)	Optical bandgap (eV)
1	−5.50	3.64	665	1.86
2	−5.48	3.61	661	1.87
3	−5.38	3.52	665	1.86
4	−5.37	3.51	664	1.86

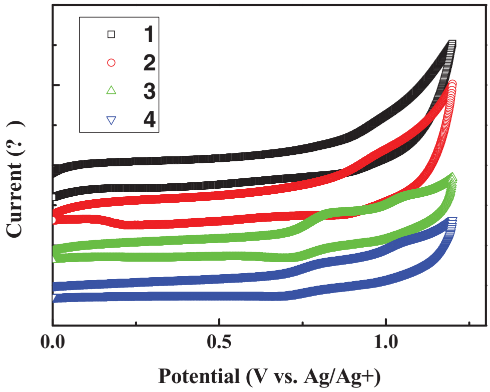


Figure 3. Cyclic voltammograms of BDT(IId)$_2$s (compound 1–4) obtained in $CHCl_3$ solution.

similar HOMO energy level.. Optical bandgaps of compound **1, 2, 3,** and **4** were measured by 1.86, 1.87, 1.86, and 1.86 eV, respectively, from UV/Vis spectroscopy. From HOMO and bandgap, LULO energy levels of compound **1, 2, 3**, and **4** were calculated by −3.64, −3.61, −3.52, and −3.51 eV, respectively. As a result, we could obtain that alkyl chains of branched or linear type has little effect of energy level control. Also, 5-(2-ethylhexyl)thien-2-yl substituent on BDT is more effective to enlarge bandgap of A-D-A type BDT(IId)$_2$ based SM donors. Finally, we can conclude bandgap can be tuned by changing the substituent on BDT, however, HOMO energy level of BDT(IId)s cannot be changed.

3.3. Morphology of BHJ Film

Figure 4 shows morphology of BHJ films and Table 2 shows detail data. Spin coated film using a mixture of compound **1** and PCBM shows best surface morphology by 0.41 nm rms

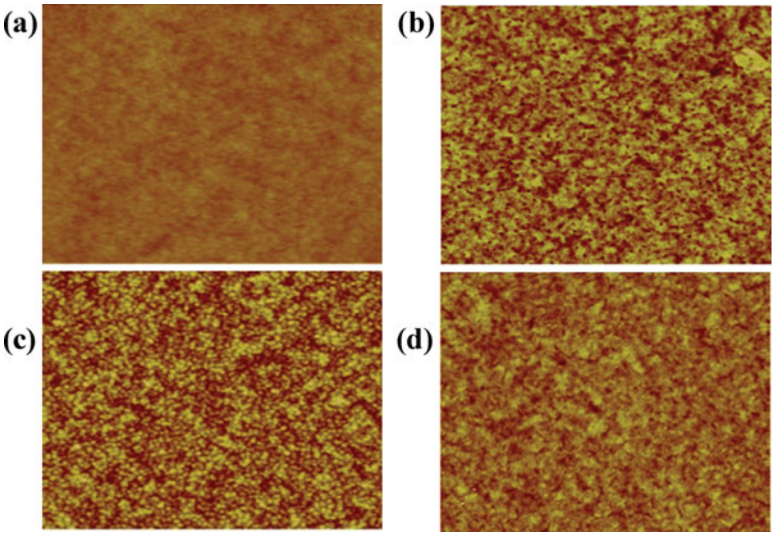


Figure 4. AFM image of active films spin coated on ITO/PEDOT:PSS substrates and foamed various donors. (a) 1. (b) 2. (c) 3,and (d) 4. The scan size for all images is 5 μm × 5 μm. The height scale for all images is 20 nm.

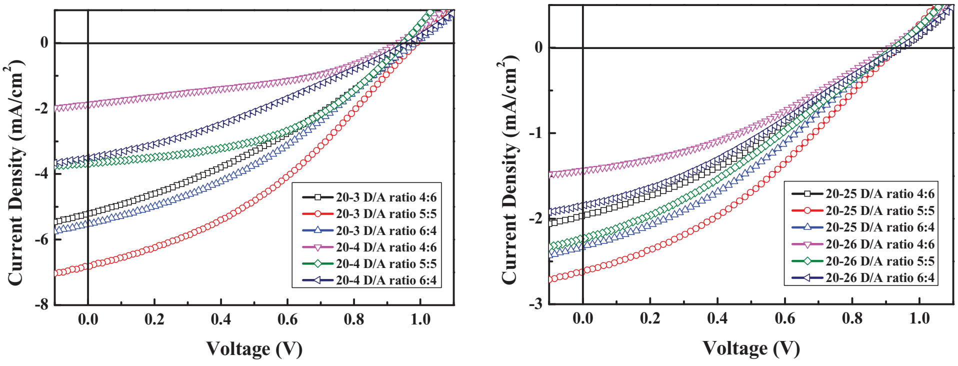
Table 2. Morphology of BHJ films with a mixture of BDT(IId)$_2$ and PCBM

Compound	Rq (nm)	Ra (nm)	rms (nm)	Rmax (nm)
1	0.47	0.37	0.41	4.17
2	2.11	1.63	2.38	24.0
3	2.16	1.71	1.95	17.5
4	1.48	1.16	1.65	27.2

and 4.17 nm Rmax. We expect that compound **1** shows best *PCE*s due to the smooth surface roughness and efficient BHJ for OPVs. 2-Ethylhexyl substituted isoindigo containing compounds (**1** and **3**) show better morphology than octyl substituted isoindigo containing compounds (**2** and **4**), that is to say, compound **3** showed higher rms roughness than compound **4**, however, compound **3** show lower Rmax than compound **4**. Thus, we can expect that branched alkyl substituted isoindigo containing compounds (**1** and **3**) show better *PCE*s than linear alkyl substituted isoindigo containing compounds (**2** and **4**).

3.4. The Performances of Organic Photovoltaic Devices

Figure 5 and Table 3 show device performance details. All of devices performance were determined at the device structure of ITO/PEDOT:PSS/BDT(IId)s:PCBM/LiF/Al. The device using compound **1** as a donor shows best performance about 2.46% of PCE with a *Voc* of 0.99 V, a *Jsc* of 6.82 mA/cm^2, and a *fill factor* of 36.6%. Compound **1** and **3** having 2-ethylhexyl substituted IId show higher photovoltaic performance than compound **2** and **4** having 1-octyl substituted IId due to the improved current density. As a result, we can conclude that branched alkyl substituent, 1-octyl group, on IId is more effective to form BHJ morphology compare to linear alkyl substituent, 2-ethylhex-1-yl. To obtain the optimum donor/acceptor ratio, we verified the mixing ratio of donor/acceptor from 6:4 to 4:6. And then we can obtain the best performance at the 5:5 ratio. At other ratio, devices



(a) (b)

Figure 5. *J-V* characteristics of ITO/PEDOT:PSS/ BDT(IId)$_2$:PCBM /LiF/Al. (a) J-V perfromance of donor using BDT substituted 5-(2-ethylhexyl)thien-2-yl. (b) J-V perfromance of donor using BDT substituted 1-ethylhexyloxy.

Table 3. Performance of OPVs with BDT(IId)$_2$ and PCBM

Compound	D/A ratio	Voc (V)	Current density (mA/Cm^2)	Fill factor (%)	PCE (%)
1	4:6	0.98	5.22	32.70	1.67
	5:5	0.99	6.82	36.57	2.46
	6:4	0.98	5.53	34.73	1.89
2	4:6	0.93	1.87	39.67	0.69
	5:5	0.95	3.67	46.13	1.61
	6:4	0.96	3.50	31.03	1.05
3	4:6	0.94	1.97	31.95	0.59
	5:5	0.93	2.62	34.59	0.85
	6:4	0.93	2.33	32.92	0.71
4	4:6	0.91	1.43	35.84	0.47
	5:5	0.93	2.23	30.76	0.64
	6:4	0.94	1.85	31.24	0.54

*Device structure: ITO/PEDOT-PSS/BDT(IId)$_2$:PCBM/LiF/Al.

performance was decreased due to current density drop, which is resulted from mismatch of hole and electron mobility balance.

4. Conclusion

We designed and synthesized BDT(IId)$_2$ based A-D-A type small molecular donor materials with variation of substituted alkyl chains for OPVs. BDT(IId)$_2$ shows definitely improved absorption property in UV region (300–400 nm), and this means BDT(IId)$_2$ derivatives can be used as small molecular donor materials for OPVs. All of device performance were determined at the device structure of ITO/PEDOT:PSS/BDT(IId)$_2$:PCBM/LiF/Al. In case of donor and acceptor ratio at 1:1 shows best device performance. Compound **1**, one of BDT(IId)$_2$, showed better *PCE* compare to other compounds (**2–4**) due to the improved photocurrent density. Especially, 2.46% of *PCE* can be obtained with a mixture of compound **1** and PCBM as a photoactive layer with a *Voc* of 0.98 V, a *Jsc* of 6.82 mA/cm^2, and a *fill factor* of 36.6%. In the future, we can report the additional donor system containing isoindigo moiety having high photovoltaic performance.

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