Toward Reliable and Accurate Evaluation of Polymer Solar Cells Based on Low Band Gap Polymers

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Toward Reliable and Accurate Evaluation of Polymer Solar Cells Based on Low Band Gap Polymers

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Along with the advances in polymer solar cells (PSCs), the accurate evaluation of novel photovoltaic polymer with various band gaps is always an important issue should be concerned, as well as need to be addressed at various research laboratories in the world. In this work, we focused on PSCs by employing some of the most efficient and well-known low band gap (LBG) polymers, for instance PBDTTT-C-T, PBDTBDD, PDPP3T, PTB7-Th, PSBTBT, PBDTTPD and obtained the corresponding spectral-mismatch factors (MMFs) under various reference cell/solar simulator combinations. Generally, there still exists ±25% spectral error even a simulator whose spectrum grade is labeled as AAA. The best way to accurately evaluate the power conversion efficiencies (*PCE*s) of LBG polymers is choosing a combination of a spectral-matched-silicon-solar-cell (match to LBG polymer's spectral responsivity spectrum) and a Class AAA solar simulator. Furthermore, our results could provide guidance for the accurate measurements of organic molecules, perovskites, and related photovoltaic technologies.

Introduction

Conjugated polymers for bulk-heterojunction polymer solar cells (PSCs) have attracted significant attention and exhibit great potential as a promising candidate for clean and renewable energy sources because of their easy structure modification, solution processability, and cheap fabrication. Research on the fabrication techniques, photovoltaic materials, device architectures and nano-scale morphology have led to impressive improvements in PSCs over the past decade and desirable power conversion efficiencies (*PCEs*) are frequently reported by various research groups[1-9]. Similar to inorganic, dye-sensitized solar cell and other photovoltaic technologies, PSCs also tend to rest heavily on its measured *PCE*[10-16]. This figure of merit has become a critical parameter for assessing the value of the PSC technology. However, several measurement errors such as light source spectral mismatch, irradiance non-uniformity and instability of light source, solar cell areas, produce great difficulties to obtain the reliable values of *PCE* for the independent laboratories around the world. Recently, the issue of erroneous efficiency reports in emerging photovoltaic technologies is emphasized in *Nature Photonics* by Henry Snaith[10] and *Solar Energy Materials & Solar Cells* by Krebs and co-workers[15]. Therefore, to provide reliable references to the PSC research, the several milestones of *PCE* in the PSC field were certificated by testing laboratories that have ISO/IEC 17025 accreditation under International Laboratory Accreditation Cooperation Mutual Recognition Arrangement (ILAC MRA) like National Renewable Energy Laboratory (NREL) in United States, the Fraunhofer Institute of Solar Energy in Germany, and the National Institute of Metrology (NIM) in China.[17-26]

The accurate measurements of organic solar cells draw attention in several research groups and they presented several useful methods for accurately measuring the efficiency since 2000.[12] As a result of continuing efforts, Yang and scientists from NREL further applied the concept of accurate measurement in PSCs and motivated the PSC field to adopt standards similar to those used for inorganic solar cells for evaluating device performance in 2006.[13] However, due to the limited photovoltaic polymers at that time, the criteria of accurate measurements like selection of reference cells or solar simulators were established on the wide band gap (WBG) polymers, such as poly(3-hexylthiophene) (P3HT) and poly[2-methoxy-5-(2′-ethylhexyoxy)-1,4-phenylene vinylene] (MEH-PPV). To achieve high efficiency, photovoltaic polymers with broader and stronger absorption spectrum are needed for well matching with the solar irradiance spectrum. **Fig. 1a** presents the UV-visible absorption spectra of P3HT and the solar irradiance spectrum. Considering that the absorption range of P3HT is 300-650 nm, the incident solar light out of this range could not be utilized efficiently, and therefore high photocurrent can be expected. During the past decade, a variety of low band gap (LBG) polymers were designed, synthesized and applied in PSC devices. Recently, PSC with record *PCE* up to 10.6% was realized by employing the newly developed LBG polymer[24]. Along with the development of PSCs, the accurate measurement and characterization of novel photovoltaic polymer with various band gaps is still an important issue should be concerned and also need to be addressed in the independent laboratories around the world. In recent works, others also emphasized the importance of device masks and solar simulators in polymer solar cells based on P3HT[14]. Any inappropriate choice of the reference cell or/and solar simulator might lead to *PCE* values departing from the true values of PSCs based on newly designed LBG polymers. To figure out the effect of reference cells and solar simulators on the spectral mismatch, novel polymers with different absorption spectrum and band gaps should be explored for accurate *PCE*s.

Although high efficiency is continually reported, the measurement details and methods are often neglected or not mentioned, particularly for newly-designed LBG polymers. In the current work, we focused on PSCs by employing some of the most efficient and well-known LBG polymers (see **Fig. 1b**) being investigated at various research laboratories in the world, for instance PBDTTT-C-T[27, 28], PBDTBDD[29, 30], PDPP3T[31, 32], PTB7-Th[33-35], PSBTBT[36], PBDTTPD[37]. Moreover, the corresponding spectral-mismatch factors (MMFs) were carefully determined under various reference cell/solar simulator combinations. Our findings indicated that most of the LBG polymers could be accurately measured under the illumination of a Class AAA solar simulator with a KG5 or KG3 filtered reference cell. However, the MMFs of the PSCs based on LBG polymers have to be further adjusted with suitable reference cells when various solar simulators are utilized. Particularly, this work revealed the critical importance of reference cells and solar simulators in reliable evaluation of device performance of LBG polymers.

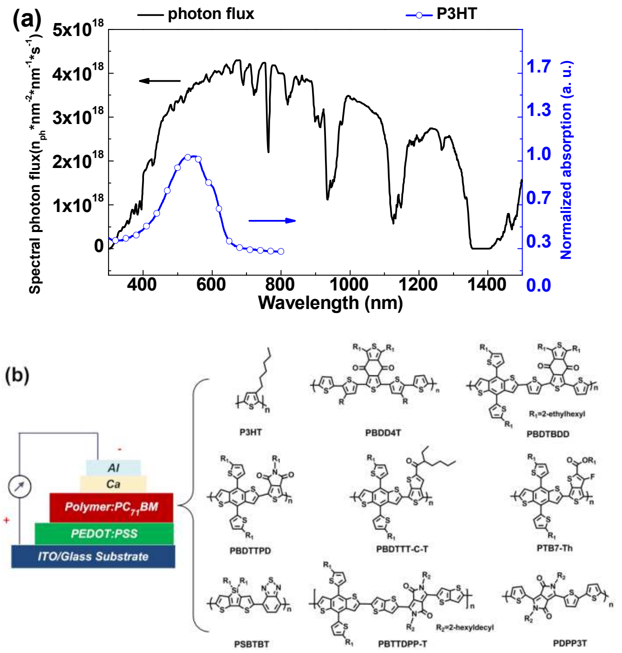


Fig. 1. (a) The spectral photon flux of AM 1.5G and the UV-vis absorption spectra of P3HT; (b) Molecular structures of photovoltaic polymers with various band gaps and device diagram of the conventional PSC.

Results and discussion

As shown in **Fig. S1**, these LBG polymers all exhibit broad absorption ranges compare with that of P3HT. These LBG polymers exhibit broad absorption ranges from 300 nm to 800 nm and some even extend to 1000 nm. Firstly, the conventional device architecture with ITO/PEDOT: PSS/polymer:PC$_{71}$BM/Ca/Al was employed to evaluate the photovoltaic performances of the LBG polymers. As described in the **Fig. 1b**, the nine different types of test PSCs had the following active layers: i) P3HT:PC$_{71}$BM; ii) PBDTTPD:PC$_{71}$BM; iii) PBDD4T:PC$_{71}$BM; iv) PBDTBDD:PC$_{71}$BM; v) PBDTTT-C-T:PC$_{71}$BM; vi) PTB7-Th:PC$_{71}$BM; vii) PSBTBT:PC$_{71}$BM; viii) PBTTDPP-T:PC$_{71}$BM; ix) PDPP3T:PC$_{71}$BM. The fabricated details of active layers are consistent with the previous works and provided in supporting information. These nine test cells almost represent the state-of-the-art types of PSCs. For comparison, the *J-V* curves (see **Fig. 2a** and **2b**) of conventional PSCs based on LBG polymers and P3HT were measured under the irradiance of a Class AAA solar simulator in ICCAS as calibrated by a silicon reference cell with KG3 filter, and the photovoltaic parameters were collected in **Table 1**. The external quantum efficiency (*EQE*) curves of the LBG polymers-based PSCs were also performed and illustrated in **Fig. 2c** and **2d**. Obviously, the spectral responsivity ranges are quite tunable for these LBG polymers due to the various band gaps (see **Table 1**). All of the LBG polymers exhibited significantly improved photovoltaic performance than P3HT. Among these LBG polymers, the ultra-small band gap polymers (E_g<1.5 eV) including PSBTBT, PBTTDPP-T, and PDPP3T delivered considerable *PCE* approaching 6~7% and PTB7-Th played leading role with *PCE* up to 8.9% in conventional PSC.

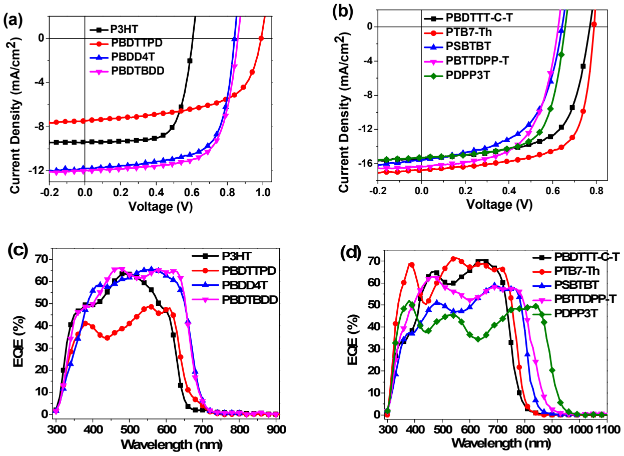


Fig. 2. (a) *J-V* curves, (b) *EQE* curves of conventional PSCs based on LBG polymers measured in our laboratory.

Then, a PSC device based on PTB7-Th/PC$_{71}$BM was randomly selected and sent to NIM for the standard measurements as calibrated by two types of reference cells. As depicted in **Fig. 3**, the open-circuit voltage (V_{oc}) and fill factor (*FF*) are identical in both tests, while the short-circuit current densities (J_{sc}) are quite different without using MMF calibrations.

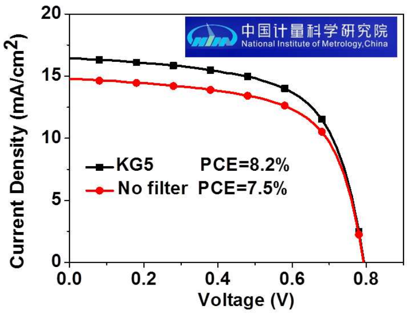


Fig. 3. The measured *J-V* results of a PTB7-Th/PC$_{71}$BM-based PSC in NIM, china.

For the accurate measurements of *PCE*s and J_{sc}s, the following essential components concerning MMF should be obtained: (i) The AM 1.5 G standard irradiance spectrum; (ii) The irradiance spectrum of solar simulator; (iii) the spectral responsivity of reference cell; and (iv) the spectral responsivity of test PSC. Since the AM 1.5G irradiance spectrum could be available from the website of NREL and the spectral responsivity of test PSC could be easy accessed by EQE measurements or from the manual. Then, the spectral responsivity of reference cell and the irradiance spectrum of solar simulator are the two essential issues in the accurate measurements of test PSCs with various spectral responsivities.

Accordingly, the effect of reference cells on MMF should be investigated in PSCs based on LBG polymers. Under the irradiance of a Class AAA solar simulator (SAN-EI Electric Co., Ltd.) in our laboratory at ICCAS (see **Fig. 4a**), aforementioned nine types of test PSCs and six types of silicon reference cells were selected to calculate MMFs for different test-cell/reference-cell combinations. The reference cells were an unfiltered silicon cell (Si for short) and filtered silicon cells respectively with a KG5 bandpass colored glass filter (KG5 for short, the same below), a KG3 bandpass colored glass filter, a BG40 bandpass colored glass filter, a 835 nm bandpass colored glass filter, and a 900 nm bandpass colored glass filter. The spectral responsivity of reference cell could be also accessed by EQE measurements or from the manual. Herein, the spectral responsivities of these reference cells were measured by NIM, as shown in **Fig. 4b**. Then, we can calculate the MMFs of these reference cell/test cell combinations with **Equation 1**.[13]

$$MMF=\frac{\int_{\lambda_1}^{\lambda_2}E_{ref}(\lambda)s_{ref}(\lambda)d\lambda\int_{\lambda_1}^{\lambda_2}E_{source}(\lambda)s_{test}(\lambda)d\lambda}{\int_{\lambda_1}^{\lambda_2}E_{ref}(\lambda)s_{test}(\lambda)d\lambda\int_{\lambda_1}^{\lambda_2}E_{source}(\lambda)s_{ref}(\lambda)d\lambda}\qquad\textbf{(Equation 1)}$$

where $E_{ref}(\lambda)$ is the spectral irradiance of AM 1.5 G reference spectrum, $E_{source}(\lambda)$ is the spectral irradiance of solar source (simulator), $s_{ref}(\lambda)$ is the spectral responsivity of the reference cell, and $s_{test}(\lambda)$ is the spectral responsivity of the test PSC, each as a function of the wavelength (λ).

As illustrated in **Fig. 5a**, most of the LBG polymers could be well certificated by KG3 or KG5 filtered silicon reference cells, affording MMFs within 1% error of unity. However, the MMFs of PSBTBT, PBTTDPP-T and PDPP3T are beyond the 1% deviation and thus these ultra-small band gap polymers need more appropriate reference cells. Alternatively, KG5, KG3 or 900 nm filtered silicon cell may be the relatively appropriate choice as reference cells for these ultra-small band gap polymers due to the relatively low deviation (<2%). Accordingly, we could rate the device performance of LBG polymers-based PSCs with MMF calibrations by the use of the KG3 filtered reference cell/Class AAA solar simulator combination. As shown in the **Table 1**, the J_{sc} and *PCE* values are calibrated with the corresponding MMFs, and PTB7-Th is still the best-performing photovoltaic polymer with *PCE* reaching 9.0%.

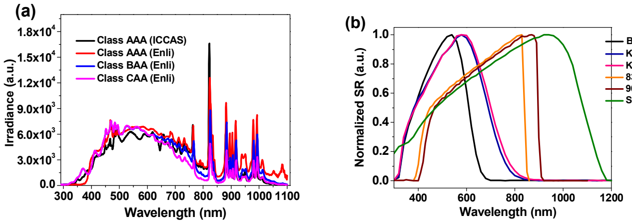
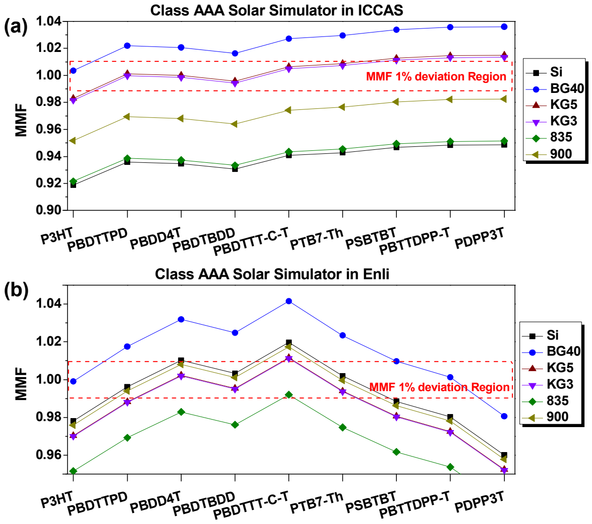


Fig. 4. (a) The irradiance spectra of various grades of used solar simulators; (b) Spectral responsivities of six different reference cells.

Solar simulators also play an equal role with reference cells in accurate measurements of *PCE*s. According to the IEC 60904-9 and ASTM E927-10 standards, spectral mismatch, irradiance spatial non-uniformity, and temporal instability are three important parameters to define a solar simulator. And each parameter is classified in one of three classes: A, B, or C. A solar simulator meeting class A specifications in all three qualifications is referred to as a Class AAA solar simulator. The first A means the spectral mismatch between AM1.5 G spectrum and irradiation spectrum of solar simulator is below 25%. That is to say, even for a simulator whose spectrum grade is labeled as AAA, there is still ± 25% error according to IEC 60904-9 standard. That should be the main reason of MMF. The irradiance spectra of various grades of solar simulators in this work are provided by a solar spectrometer (Enli tech, SS-E1000) which was calibrated and traceable to SI unit. Then the above reference cell/test cell combinations were also calculated by the use of a Class AAA solar simulator (Enli Tech Co., Ltd.) in Enli with a different irradiance spectra (see **Fig. 4b**). Although the MMFs are quite similar to that determined from ICCAS Class AAA solar simulator for a variety of reference cell/test cell combinations, the MMFs are quite different for several combinations. For instance, the MMF of PDPP3T-based PSC/KG3 filtered reference cell combination is calculated to be 1.01 with the Class AAA solar simulator in ICCAS and 0.96 by the use of the Class AAA solar simulator in Enli. The MMF of PBTTDPP-T-based PSC/BG40 filtered reference cell combination is 1.00 with the Class AAA solar simulator in Enli and 1.04 by the use of the Class AAA solar simulator in Enli. Besides, we also calculated the MMFs of these reference cell/test cell combinations by using a Class BAA and a Class CAA solar simulator (Enli Tech Co., Ltd.) in Enli, as shown in **Fig. 5c and 5d**. Some of the LBG polymers still could be accurately measured by carefully selecting the specific reference cells when Class BAA or CAA solar simulators are utilized. It could be observed that Class BAA and especially Class CAA solar simulator fail to certificate the PSCs based on PTB7-Th, PSBTBT, and PBTTDPP-T due to the high deviations ranging from 25% to 50%, which might lead to severe deviations to the true values. Incorrect spectral mismatch should be also avoided introducing in the reported *PCE*s. Nevertheless, for the PSCs based on polymers with optical band gap low than 1.4 eV (i.e., PDPP3T), more suitable reference cell are still needed to be explored when the irradiance spectra of solar simulator are varied. Clearly, MMF of a typical reference cell/test cell combination varied with the irradiance spectrum of solar simulators and should be noticed for a specific measurement of *J-V* characteristics.



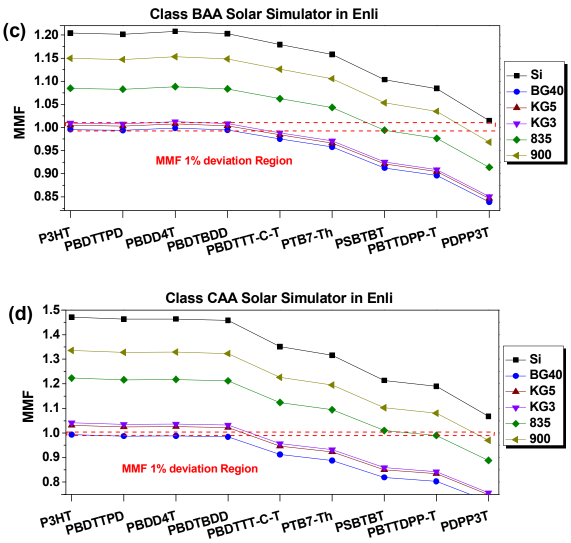


Fig. 5. The calculated MMFs of photovoltaic polymers with different band gaps by the use of Class AAA solar simulator in ICCAS (a), and Class AAA (b), Class BAA (c), and Class CAA (d) solar simulators in Enli.

Table 1. Photovoltaic properties of donor polymers and their corresponding best-performance device performances under AM1.5G 100mW/cm^2 using a KG3 filtered reference cell/Class AAA solar simulator combination in ICCAS.

Polymers	E_g^{opt} [eV]	V_{OC} [V]	*FF*	J_{SC} [mA/cm^2]	*PCE*^{a} [%]	*PCE*^{b} [%]
P3HT	1.91	0.60	0.70	9.4	3.95	3.87
PBDTTPD	1.82	0.99	0.59	7.5	4.38	4.38
PBDD4T	1.77	0.84	0.68	11.8	6.74	6.74
PBDTBDD	1.77	0.86	0.68	12.0	7.02	7.02
PBDTTT-C-T	1.58	0.77	0.65	15.3	7.66	7.74
PTB7-Th	1.58	0.78	0.68	16.8	8.91	9.00
PSBTBT	1.45	0.64	0.56	15.6	5.60	5.67
PBTTDPP-T	1.43	0.63	0.60	16.4	6.20	6.26
PDPP3T	1.33	0.66	0.67	15.3	6.77	6.84

a. *PCE* without MMF calculations
b. *PCE* with MMF calculations

Overall, a general method and various examples to accurately measure the photovoltaic parameters especially *PCE* of LBG polymers under standard test conditions are presented. For a device measurement laboratory, a general set of procedures of reliable efficiency measurements of PSCs could be as follows: (i) measure the EQE curves of test cell; (ii) obtain the spectral responsivity of the reference cell from manual or from EQE measurement; (iii) obtain the irradiance spectra of the solar simulator by a irradiance spectrometer; (iv) calculate the MMF of the test cell; (v) measure the test cell and calculate the true *PCE* by using MMF calibration. Notably, for a specific solar simulator in independent laboratories, any inappropriate choice of the reference cell might lead to *PCE* values deviating from the true values of PSCs based on newly designed LBG polymers with various band gaps or absorption ranges. In addition, the inaccurate calculation of device area also affects the true *PCE*s and the effect of device area should be regarded as an important aspect of accurate measurements[14,15], which is out of the scope of this study. This issue might be avoided by using aperture with appropriate size, which can be calibrated by a sophisticated microscopy.

Experimental

Materials. P3HT, PBDTTT-C-T, PBDTTPD, PDPP3T, PSBTBT and PTB7-Th were purchased from Solarmer Energy Inc. PBDTBDD[29], PBDD4T[38], and PBTTDPP-T[39] was synthesized in our laboratory following the previous literatures. The PEDOT:PSS (Heraeus Clevios™ P VP AI 4083) and electrode materials are commercial available products. The other chemicals are commercial available products and used without any further purification. **Fabrication and Characterization of PSC devices.** The general device fabrication details of these polymers are described elsewhere[40] and also provided in **Table S1**. The various types of silicon reference cells and solar simulators are provided by Enli Technology Co. Ltd. The current density-voltage (*J-V*) characteristics were measured in Institute of Chemistry, Chinese Academy of Sciences (ICCAS) under an Keithley 2400 Source Measure Unit under by the Class AAA XES-70S1 solar simulator (SAN-EI Electric Co., Ltd.) equipped with an AM 1.5G filter at a calibrated light intensity of 100 mW/cm^2. And the standard calibrations of reference cells were conducted in the optics division of NIM. The *EQE* data were measured, by the use of the Solar Cell Spectral Response Measurement System (QE-R3011, Enli Technology Co. Ltd.), which was calibrated with a silicon reference solar cell calibrated by NIM. During the measurements, a shadow mask with a single aperture (4.15 cm^2) was placed onto the PSCs in order to define the photoactive area. The areas of PSC and shadow mask were calibrated by the optical microscope and also calibrated by the length division of NIM.

Conclusions

In the current work, we focused on the accurate measurements of PSCs based on some of the most efficient and well-known LBG polymers such as PDPP3T， PSBTBT， PBDTTPD， PTB7-Th and a general set of procedures were provided to obtain reliable *PCE* results for LBG polymers. Six reference cells and four solar simulators with different classifications were selected for calculating MMFs of nine types of LBG polymers-based PSCs with respect to the AM 1.5G reference spectrum. Specifically, the device performance of PSCs based on most of LBG polymers could be well evaluated by a spectral-matched-silicon reference cell with a Class AAA solar simulator. Our results clearly indicated that choosing an appropriate reference cell and solar simulator is of great significance in the measurements of PSCs based on newly designed LBG polymers. More importantly, the results based on LBG polymers with various band gaps could also provide guidance for the accurate measurements of the emerging hybrid organic/inorganic perovskite solar cells due to their tunable band gaps.

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Notes and references

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† Electronic Supplementary Information (ESI) available: Additional device fabrication details of LBG polymer/PCBM blends, and Absorption spectrum of different photovoltaic polymers. See DOI: 10.1039/b000000x/

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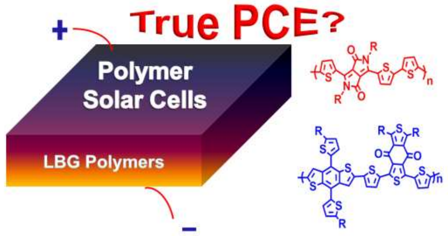
Table of Content



To accurately evaluate the power conversion efficiencies, a general set of procedures were provided for low band gap polymers.