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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 polymers with various band gaps is an important issue that should be concerned, as well as needs to be addressed at various research laboratories in the world. In this work, we have 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 and PBDTTPD, and obtained the corresponding spectral-mismatch factors (MMFs) under various reference cell/solar simulator combinations. Generally, there still exists $\pm 25\%$ spectral error even for a simulator whose spectrum grade is labeled as AAA. The best way to accurately evaluate the power conversion efficiencies (PCEs) of LBG polymers is by 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.

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Introduction

Conjugated polymers for bulk-heterojunction polymer solar cells (PSCs) have attracted significant attention and exhibit immense potential as a promising candidate for clean and renewable energy sources because of their easy structural 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 cells and other photovoltaic technologies, PSCs also tend to rest heavily on their 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 the light source, and solar cell areas produce huge difficulties to obtain the reliable values of PCE for independent laboratories around the world. Recently, the issue of erroneous efficiency reports in emerging

photovoltaic technologies has been emphasized in Nature Photonics by Henry Snaith¹⁰ and Solar Energy Materials & Solar Cells by Krebs and co-workers.¹⁵ Therefore, to provide reliable references to the PSC research, 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) such as the National Renewable Energy Laboratory (NREL) in the 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 among several research groups, and a number of useful methods have been presented for accurately measuring the efficiency since 2000.¹² As a result of continuing efforts, in 2006, 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 the device performance.¹³ However, due to limited photovoltaic polymers at that time, the criteria of accurate measurements, such as the selection of reference cells or solar simulators, were established on wide band gap (WBG) polymers such as poly(3-hexylthiophene) (P3HT) and poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene vinylene] (MEH-PPV). To achieve high efficiency, photovoltaic polymers with broader and stronger absorption spectrum are needed for matching well 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

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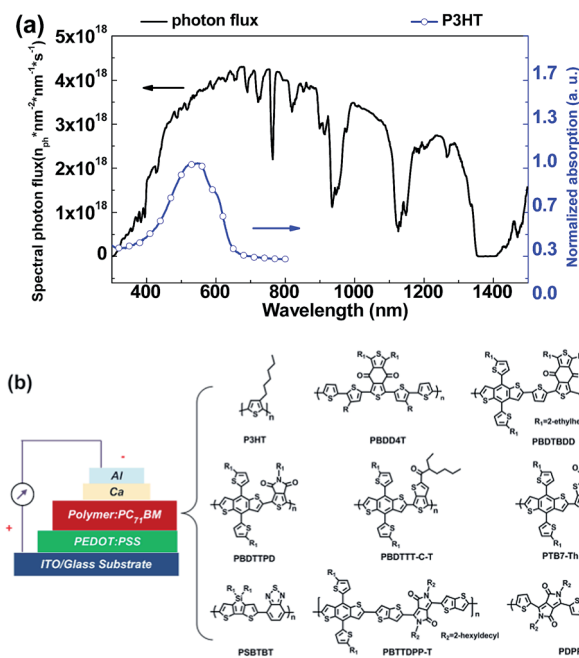


Fig. 1 (a) The spectral photon flux of AM 1.5G and the UV-vis absorption spectra of P3HT; (b) molecular structures of photovoltaic polymers with various band gaps and device diagram of the conventional PSC.

range could not be utilized efficiently, and therefore a high photocurrent cannot be expected. During the past decade, a variety of low band gap (LBG) polymers were designed, synthesized and applied in PSC devices. Recently, PSCs with record PCE up to 10.6% were realized by employing the newly developed LBG polymer.²⁴ Along with the development of PSCs, the accurate measurement and characterization of novel photovoltaic polymers with various band gaps is still an important issue that should be concerned and also needs to be addressed in the independent laboratories around the world. In recent works, other groups have 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 spectra and band gaps should be explored for accurate PCEs.

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 have 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} PSBTBT³⁶ and PBDTPD.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. In particular, this work revealed the critical importance of reference cells and solar simulators in the reliable evaluation of the device performance of the LBG polymers.

Results and discussion

As shown in Fig. S1,[†] all these LBG polymers exhibit broad absorption ranges compared with that of P3HT. These LBG polymers exhibit broad absorption ranges from 300 nm to 800 nm and some even extend to 1000 nm. First, the conventional device architecture with ITO/PEDOT:PSS/polymer:PC₇₁BM/Ca/Al was employed to evaluate the photovoltaic performances of the LBG polymers. As described in Fig. 1b, the nine different types of test PSCs had the following active layers: (i) P3HT:PC₇₁BM; (ii) PBDTPD:PC₇₁BM; (iii) PBDD4T:PC₇₁BM; (iv) PBDTBDD:PC₇₁BM; (v) PBDTTT-C-T:PC₇₁BM; (vi) PTB7-Th:PC₇₁BM; (vii) PSBTBT:PC₇₁BM; (viii) PBTTDPP-T:PC₇₁BM; (ix) PDPP3T:PC₇₁BM. The fabricated details of active layers are consistent with previous works and are provided in the ESI.[†] These nine test cells almost represent the state-of-the-art types of PSCs. For comparison, the *J*-*V* curves (see Fig. 2a and b) 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 are illustrated in Fig. 2c and d. Evidently, the spectral responsivity ranges are quite tunable for these LBG polymers due to various band gaps (see Table 1). All of the LBG polymers exhibited significantly improved photovoltaic performance compared to P3HT. Among these LBG polymers, the ultra-small band gap polymers ($E_g < 1.5$

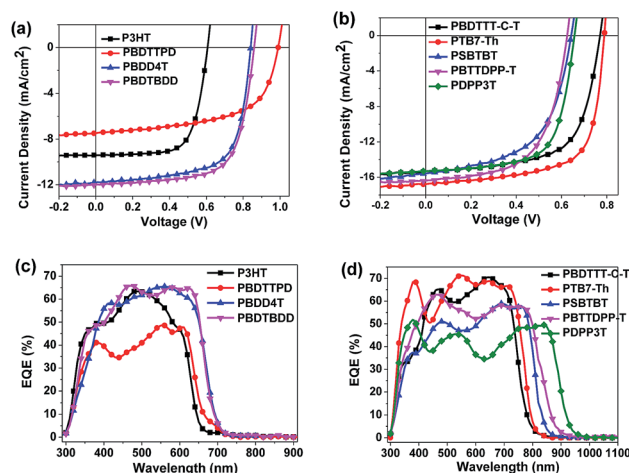


Fig. 2 (a) *J*-*V* curves, (b) EQE curves of conventional PSCs based on LBG polymers, measured in our laboratory.

Table 1 Photovoltaic properties of donor polymers and their corresponding best-performance device performances under AM 1.5G 100 mW 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
PBTDDPP-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.

eV), including PSBTBT, PBTDDPP-T, and PDPP3T, delivered considerable PCE approaching 6–7%, and PTB7-Th played the leading role with PCE up to 8.9% in a conventional PSC.

Then, a PSC device based on PTB7-Th/PC₇₁BM 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.

For the accurate measurements of PCEs and J_{sc} s, the following essential components concerning MMF should be obtained: (i) the AM 1.5G standard irradiance spectrum; (ii) the irradiance spectrum of the solar simulator; (iii) the spectral responsivity of the reference cell; and (iv) the spectral responsivity of the test PSC. The AM 1.5G irradiance spectrum could be obtained from the website of NREL, and the spectral responsivity of the test PSC could be easily accessed by EQE measurements or from the manual. Then, the spectral responsivity of the reference cell and the irradiance spectrum of the 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), the 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 used were an unfiltered silicon cell (abbreviated to Si) and filtered silicon cells, respectively, with a KG5 bandpass colored glass filter (abbreviated to KG5), 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 the reference cell could also be 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 eqn (1).¹³

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

where $E_{\text{ref}}(\lambda)$ is the spectral irradiance of the AM 1.5G reference spectrum, $E_{\text{source}}(\lambda)$ is the spectral irradiance of the solar source (simulator), $s_{\text{ref}}(\lambda)$ is the spectral responsivity of the reference cell, and $s_{\text{test}}(\lambda)$ 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, PBTDDPP-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 cells may be a 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%.

Solar simulators also play an equivalent role with reference cells in the accurate measurements of PCEs. 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. Moreover, each parameter is classified in one of the three classes: A, B, or C. A solar simulator meeting class A specifications in all

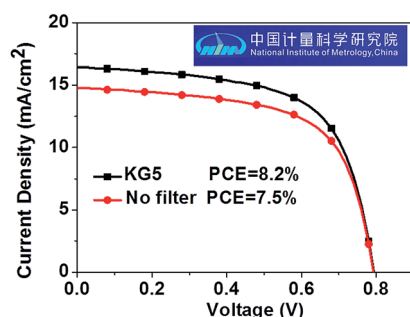


Fig. 3 The measured J - V results of a PTB7-Th/PC₇₁BM-based PSC in NIM, china.

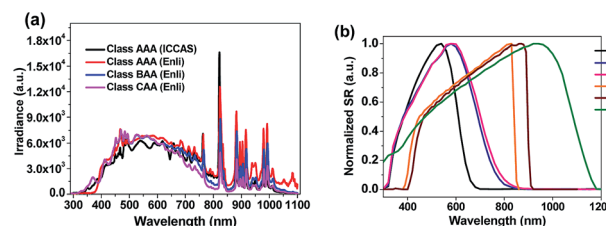


Fig. 4 (a) The irradiance spectra of various grades of used solar simulators; (b) spectral responsivities of six different reference cells.

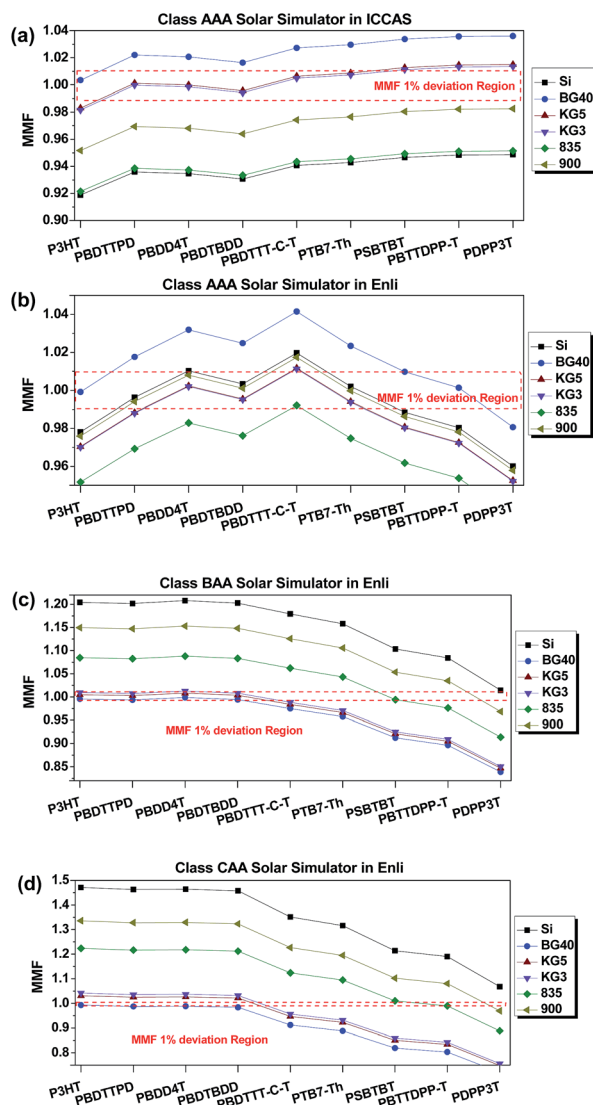


Fig. 5 The calculated MMFs of photovoltaic polymers with different band gaps by the use of a Class AAA solar simulator in ICCAS (a), and Class AAA (b), Class BAA (c), and Class CAA (d) solar simulators in Enli.

three qualifications is referred to as a Class AAA solar simulator. The first A means that the spectral mismatch between the AM 1.5G spectrum and the irradiation spectrum of the solar simulator is below 25%. In other words, even for a simulator whose spectrum grade is labeled as AAA, there is still $\pm 25\%$ error according to the IEC 60904-9 standard. This should be the main reason for 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 units. Then, the abovementioned 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 spectrum (see Fig. 4b). Although the MMFs are quite similar to that determined from the 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 the 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 the 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 ICCAS. In addition, we also calculated the MMFs of these reference cell/test cell combinations using a Class BAA and a Class CAA solar simulator (Enli Tech Co., Ltd.) in Enli, as shown in Fig. 5c and d. Some of the LBG polymers could still be accurately measured by carefully selecting the specific reference cells when Class BAA or CAA solar simulators were utilized. It could be observed that Class BAA and, especially, Class CAA solar simulators 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 from the true values. Obviously, incorrect spectral mismatch of novel photovoltaic polymer should be also avoided introducing in the reported PCE when BAA or CAA solar simulator is selected. Nevertheless, for the PSCs based on polymers with optical band gaps lower than 1.4 eV (*i.e.*, PDPP3T), more suitable reference cells are still needed to be explored when the irradiance spectra of solar simulator are varied. Clearly, the 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.

Overall, a general method and various examples to accurately measure the photovoltaic parameters, especially the 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 the test cell; (ii) obtain the spectral responsivity of the reference cell from the manual or from EQE measurements; (iii) obtain the irradiance spectra of the solar simulator by an irradiance spectrometer; (iv) calculate the MMF of the test cell; (v) measure the test cell and calculate the true PCE using MMF calibration. Notably, for a specific solar simulator in an independent laboratory, any inappropriate choice of the reference cell might lead to PCE values deviating from the true values of PSCs based on the newly designed LBG polymers with various band gaps or absorption ranges. In addition, the inaccurate calculation of device area also affects the true PCEs, 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 the use of an aperture with appropriate size, which can be calibrated by 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-T³⁹ were synthesized in our laboratory, following previous literature. The PEDOT:PSS (Heraeus Clevis™ P VP AI 4083) and electrode materials are commercially available products. Other chemicals are also commercially available products and are used without any further purification.

Fabrication and characterization of PSC devices

The general device fabrication details of these polymers are described elsewhere⁴⁰ 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 the Institute of Chemistry, Chinese Academy of Sciences (ICCAS) under a Keithley 2400 Source Measure Unit 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} . Moreover, the standard calibrations of reference cells were conducted in the optics division of NIM. The EQE data were measured using 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 mm^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 have focused on the accurate measurements of PSCs based on some of the most efficient and well-known LBG polymers such as PDPP3T, PSBTBT, PBDTPD and PTB7-Th, and a general set of procedures has been provided to obtain reliable PCE results for the 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 the 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 huge significance in the measurements of PSCs based on the newly designed LBG polymers. 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.

Acknowledgements

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