ADVANCED MATERIALS

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Conjugated Polymer Photovoltaic Materials with Broad Absorption Band and High Charge Carrier Mobility**

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Polymer solar cells (PSCs) have attracted great attention in recent years because of their advantages of easy fabrication, low cost, light weight, and potential for flexible devices. However, the power conversion efficiency (PCE) of the PSCs needs to be improved

 $\begin{array}{c} 1.0 \\ 0.8 \\ 0.0 \\ 0.6 \\ 0.2 \\ 0.2 \\ 0.3 \\ 0.0 \\$

for future commercial applications. Factors limiting the PCE of the PSCs include the low exploitation of sunlight due to the narrow absorption band of conjugated polymers, and the low charge-transport efficiency in the devices due to the lower charge-carrier mobility of the polymer photovoltaic materials. In this Research News article, recent progress in new conjugated polymer photovoltaic materials synthesized by our group and others is reviewed, including polythiophene (PT) and poly(thienylene vinylene) derivatives with conjugated side chains for a broad absorption band, crosslinked PT derivatives with conjugated bridges for higher hole mobility, and low-bandgap donor-acceptor copolymers for broad, red-shifted absorption to match the solar spectrum.

1. Introduction

Harvesting energy directly from sunlight using photovoltaic technology is increasingly recognized around the world as a critical hi-tech solution to the growing energy crisis. Polymer solar cells (PSCs) have attracted considerable attention in recent years because of their unique advantages, such as low cost, light weight, solution processibility, and flexibility.^[1-4] However, the power conversion efficiency (PCE) of the PSCs needs to be improved for future commercial applications.

PSCs are commonly composed of a photoactive blend layer of conjugated polymer (as donor) and PCBM ([6,6]-phenyl- C_{61}

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The factors limiting the PCE of the PSCs include the low exploitation of the sunlight due to the narrower absorption band of the absorption spectra of conjugated polymers in comparison with the solar spectrum and the mismatch of the two spectra, and the low charge transport efficiency in the



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devices due to the low charge carrier mobility of polymer photovoltaic materials. Therefore, much effort has been devoted to improving the absorption spectra and charge carrier mobility of conjugated polymer photovoltaic materials. The strategies used to date in designing new photovoltaic polymers involve attaching conjugated side chains to the polymer main chains to broaden the absorption band,^[5-10] crosslinking of the polymer main chains with conjugated bridges to enhance the charge carrier mobility,^[11–13] and copolymerization of various electron-rich (donor) and electrondeficient (acceptor) units to produce lower bandgap polymers and thus red-shift the absorption of the conjugated polymers.^[14-24] Figure 1 shows the molecular structures of conjugated polymer photovoltaic materials, including poly(3hexylthiophene) (P3HT), which is a traditional and most representative photovoltaic polymer donor. In the following, the progress of the design and synthesis of new conjugated polymer photovoltaic materials by our group and others is reviewed and discussed.

2. Polythiophenes and Poly(thienylene vinylene)s with Conjugated Side Chains

It is well known that extension of conjugation degree leads to enhancement and red-shift of the absorption spectra of conjugated polymers. The most efficient polymer photovoltaic material is the polythiophene (PT) derivative P3HT.^[2,3] Based on this consideration, we have designed and synthesized PT derivatives with conjugated phenylenevinylene side chains (polymers 1–4 in Fig. 1)^[5] to extend the conjugation of PTs. The first polymer we synthesized is 1. Interestingly, the polymer shows two absorption peaks in the UV-vis region (see Fig. 2a); the absorption peak in the UV region is ascribed to the absorption of the thiophene units with the conjugated side chains and that in the visible region corresponds to the absorption of the PT main chains. The weak visible absorption of 1 results from the distortion of the polymer main chains by the conjugated side chains. Then polymers 2-4 were synthesized, aimed at strong and broad absorption of the polymers. By doubling the conjugation length of the side chains in 3 and 4, we were able to red-shift the UV absorption from 328 nm to 371 nm, and by controlling the ratio of the thiophene units with the conjugated side chains in 2 and 4, the visible absorption was enhanced and red-shifted. After the structure modification, polymer 4 showed a broad absorption in the wavelength range from 300 nm to 650 nm with a valley at around 450 nm.^[5]

In order to further improve the visible absorption and intensify the absorbance at around 450 nm of 4, we synthesized PT derivatives with conjugated bi(thienylenevinylene) side chains (6-8),^[6] instead of the bi(phenylenevinylene) side chains in 4, in order to red-shift the UV absorption of the polymers to the visible region, because poly(thienylenevinylene) (PTV) is a low-bandgap polymer. The result confirmed our expectations. Polymers 6–8, especially polymer 8, display a strong and broad absorption plateau in the visible region from

350 nm to 650 nm (see Fig. 2b). In addition, the HOMO energy level of the PTs with bi(thienylenevinylene) conjugated side chains dropped by ca. 0.2 eV in comparison with that of P3HT without the conjugated side chains, which could lead to higher open circuit voltage of the PSCs using the polymer as donor.

Among the PT derivatives with the thienylenevinylene conjugated side chains, polymer **5** shows a distinct thermochromic characteristic.^[7] As shown in Figure 3, the absorption spectrum of polymer **5** film was red-shifted by ca. 100 nm after thermal annealing at 130°C for 10 min, showing a very broad absorption bandwidth ranging from 300 nm to 700 nm. The bandgap of the polymer **5** film is reduced from 2.03 eV to 1.77 eV. X-Ray diffraction (XRD) shows that the heat treatment helps the polymer chains to realign and improves coplanarity of the polymer chains.^[7]

The photovoltaic properties of the polymers were investigated by fabricating bulk-heterojunction polymer solar cells based on a blend of the polymer as donor and PCBM as acceptor with a weight ratio of 1:1. PSC of the device based on **8** reached 3.18% under AM1.5, 100 mW/cm² illumination, which is a 38% increase compared to that of the device based on P3HT under the same experimental conditions.^[6] The IPCE (input photon to converted current efficiency) results of the devices revealed that the broad absorption in the visible region contributes to the higher efficiency of the device based on **8**. The results indicate that the strategy for broad absorption involving attaching conjugated side chains is efficient in improving the PCE of the PSCs.

In pursuing more broad absorption conjugated polymers in the visible region, we further synthesized three poly(thienylenevinylene) (PTV) derivatives with bi(thienylenevinylene) conjugated side chains (14-16).^[8] Indeed, the PTV derivatives show strong and very broad absorption in the visible region (see Fig. 2c). Especially polymer 16 displays two absorption peaks mixed together, showing a distinct, strong absorption plateau covering the whole visible region from 350 nm to 780 nm.^[8] Such a broad absorption band is very unusual for organic polymers. Although the photovoltaic properties of the PTVs were poor (PCE of the PSC based on 16 was 0.32% under AM 1.5, $100 \,\mathrm{mW \, cm^{-2}}$ illumination), due probably to their nonluminescent characteristic, the PCE of the PSC based on polymer 16 is increased by 52% in comparison with that of the device based on 13 without the conjugated side chains under the same experimental conditions.^[8]

On the basis of the above findings, we further synthesized PT derivatives with other conjugated side chains, such as terthiophene-vinylene $(9-11)^{[9]}$ and phenothiazine-vinylene (12).^[10] These polymers also possess a broad absorption band in the visible region. The PCE of the PSC based on 10 as donor reached 1.91% under AM 1.5, 100 mW cm⁻² illumination.^[9] In addition, polymers 10 and 12 possess high hole mobilities. The maximum hole mobility of polymer 10, measured by the space-charge-limited current (SCLC) method, reached $6.35 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is one order of magnitude higher than common PTs without the conjugated side chains $(10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$. The hole mobility of polymer 12, measured



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P(C₄H₉)₃

31

10H21

C12H25

Ċ

33

12H25 Ċ₁₀H₂₁



a) Polythiophene and poly(thienylene vinylene) derivatives with conjugated side

Figure 1. Molecular structures of photovoltaic conjugated polymers.

C8H17 C8H17

29

RRC

R=2-ethylhexyl

32

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C₈H₁₇ C₈H₁₇

30



Figure 2. Absorption spectra of the polymer films: a) PT derivatives with phenylenevinylene conjugated side chains; b) PT derivatives with bi(thienylenevinylene) conjugated side chains; c) poly(thienylenevinylene) derivatives with bi(thienylenevinylene) conjugated side chains; d) low-bandgap polymer 33.

by the SCLC method, reached $4.7 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is about two orders of magnitude higher than that of the common PTs.^[10] The field effect hole mobility of polymer **12** was as high as $6.8 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ with an on/off ratio of 2.5×10^4 , indicating that polymer **12** could also be a promising conjugated polymer material for field-effect transistors (FETs).^[10]

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The thermal stability of the polymers with the conjugated side chains was also studied by thermogravimetric analysis



Figure 3. Absorption spectra of polymer 5 film before and after thermal treatment.

(TGA). The onset decomposition temperatures of all the polymers are higher than 300°C, indicating that the thermal stability of the polymers is enough for application in optoelectronic devices.

3. Crosslinked PT Derivatives with Conjugated Bridges

Higher charge carrier mobilities are pursued by researchers in the design and synthesis of new conjugated polymers for applications in PSCs and polymer FETs, etc. As we know, the transport processes of charge carriers in conjugated polymers include charge transport along the conjugated main chains and hopping between two adjacent main chains; the interchain hopping is the limiting step of the charge transport. In order to improve the interchain charge transport for higher hole mobility, we designed and synthesized a series of crosslinked PT derivatives (**17–25**) with conjugated bridges, such as vinylene-phenylene-vinylene (VPV),^[11] vinylene-terthiophene-vinylene (VTThV),^[12] and vinylene-bi(thienylenevinylene) (VbiTV).^[13] Table 1 lists the hole mobilities of the crosslinked PTs. It can be seen that the hole mobility of polymer **19** with 5% VPV conjugated bridges reached $7 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (SCLC method).^[11] Obviously, a suitable



Table 1. Hole	mobility of the a	crosslinked polythiophen	e derivatives with conjugated b	ridges.
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Polymer Bridge		Content of bridge [%]	Mobility [cm ² V ⁻¹ s ⁻¹]	Measurement method	Ref. [11]	
17	VPV 0		5.23×10^{-6}	SCLC		
18	VPV	2	1.28×10^{-4}	SCLC	[11]	
19	VPV	5	7.01×10^{-3}	SCLC	[11]	
20	VPV	10	$2.34 imes 10^{-5}$	SCLC	[11]	
21	VTThV	2	4.70×10^{-3}	SCLC	[12]	
22	VTThV	4	$2.58 imes 10^{-3}$	SCLC	[12]	
23	VTThV	8	$9.48 imes 10^{-4}$	SCLC	[12]	
24	VbiTV	2	1.1×10^{-3}	FET	[13]	
25	VbiTV	4	$3.8 imes10^{-3}$	FET	[13]	

content of conjugated bridges in the crosslinked PTs leads to a dramatic increase of the hole mobility of the polymers. However, too high a percentage of crosslinking resulted in poor solubility and a decrease of the hole mobility, due probably to the distortion of the polymer main chains. The crosslinking with VTV bridges caused a blue shift of the absorption of the polymer films,^[11] which is a weak point for the application as photovoltaic materials. The hole mobility of polymer 21 with 2% VTThV conjugated bridges is about two orders of magnitude higher than that of the corresponding polymer 17 without the conjugated bridges.^[12] Compared with the crosslinked PTs with VPV conjugated bridges, the longer VTThV conjugated bridge influenced the absorption spectra of the crosslinked polymers very little (slight blue-shift), which may be related to less distortion of the main chains of the polymers with the longer VTThV conjugated bridges. The power conversion efficiency of the PSC based on the blend of 21 and PCBM (1:1, w/w) reached 1.72% with shortcircuit current density J_{sc} of 6.8 mA cm⁻² under AM 1.5, $100 \,\mathrm{mW \, cm^{-2}}$, illumination, which is two times that of the device based on polymer 17.^[12] For polymers 24 and 25 with VbiTV conjugated bridges, FET behavior was studied. The FET hole mobility of 25 reached 3.8×10^{-3} cm² V⁻¹ s⁻¹ with an on/off ratio of 10⁴ and threshold voltage about 24 V.^[13]

The results indicate that high hole mobility of conjugated polymers can be obtained from conjugated crosslinked polymers with a suitable content of conjugated bridges. The conjugated crosslinked structure provides an effective route to improving the charge mobility, which results in higher PCE of PSCs.

4. Low-Bandgap Polymers

In the design and synthesis of high efficiency polymer photovoltaic materials, low-bandgap conjugated polymers have been broadly pursued in recent years, in order to match the absorption spectra of the polymers to the solar spectrum. The peak photon intensity of the solar spectrum is at ca. 700 nm. To make the absorption spectra of the conjugated polymers match the solar spectrum, their absorption maximum should be at ca. 700 nm, which means that the bandgap (E_{o}) of the conjugated polymers should be lower than 1.74 eV.^[14] The most successful approach to achieving low-bandgap polymers is a copolymerized D–A structure.^[15-24] The copolymerization of the donor (electron-rich) with higher HOMO energy level and the acceptor (electron deficient) with lower LUMO energy level results in a lower-bandgap polymer due to an intrachain charge transfer from donor to acceptor. To date the most successful low-bandgap polymer photovoltaic materials are copolymers of benzothiadiazole (or analogues) as the acceptor with fluorene or fluorene-like units (replacement of two benzene rings by two thiophene rings, or replacement of 9-C by Si or N) as the donor (26–30). Table 2 lists the optical bandgap, HOMO and LUMO energy levels, and photovoltaic properties of the low-bandgap polymers.

A highly efficient low-bandgap photovoltaic polymer is poly[$\{2,6-(4,4-bis(2-ethylhexyl))-4H$ -cyclopenta[2,1-b;3,4-b'] dithiophene}-*alt*- $\{4,7-(2,1,3-benzothiadiazole)\}$] (**26**) with a bandgap of 1.46 eV.^[15] The PSC based on **26** as donor blended with PCBM as acceptor (1:1 w/w) showed a PCE of 2.7% with photocurrent response extending to wavelengths longer than

Polymer	Eg[a] [eV]	E _{номо} [eV]	E _{LUMO} [eV]	V _{oc} [V]	$\int_{\rm sc}$ [mA cm ⁻²]	FF[b]	PCE [%]	Ref.
26	1.46	-5.3	-3.57	0.62	16.2	0.55	5.5	[15]
27	1.98	-	-	1.04	4.66	0.46	2.2	[17]
28	1.78	-5.14	-3.36	0.76	4.31	0.486	1.6	[18]
29	1.82	-5.39	-	0.90	9.5	0.507	5.4	[19]
30	1.88	-5.5	-3.6	0.89	6.92	0.63	3.6	[20]
31	1.85	-5.37	-3.14	0.82	13.1	0.37	4.1	[21]
32	1.20	-	-	0.56	3.1	0.58	1.1	[22]
33	1.46	-5.9	-3.9	0.63	4.2	0.39	1.0	[23]

Table 2. Electronic energy levels and optimized photovoltaic performance of some low-bandgap polymers.

[a]Optical energy gap determined from the absorption edge of the polymer film. [b]Fill factor.

5



900 nm.^[15] This is the first low-bandgap polymer with highly efficient photovoltaic conversion extending into the IR spectral region. Using the C_{70} derivative of PCBM leads to a further higher efficiency of 3.2% for the device based on **26**,^[15] because the visible absorption of C_{70} is stronger than that of C_{60} . The excellent performance of polymer **26** can be attributed to its broad absorption spectrum and high hole mobility $(2 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$, FET method), as well as the superior miscibility of **26** with PCBM. Recently Heeger and coworkers further increased the PCE to 5.5% by using alkanedithiols as a solvent additive to improve the blend morphology.^[16]

Andersson and coworkers first reported copolymer 27 with a photovoltaic efficiency of 2.2%, benefiting from its broad absorption and possibly high mobility.^[17] Since then, the alternative copolymers of fluorene and acceptor units have drawn much attention as high efficiency photovoltaic polymers. A closely related polymer (28) with O-alkyl side chains on the thiophene rings of 27 was synthesized by Pei and coworkers.^[18] PCE of 1.6% was obtained for the device with 28 as donor blended with PCBM (1:4, w/w) (AM 1.5 G, $80 \,\mathrm{mW \, cm^{-2}}$). Very recently, Cao and coworkers reported a new low-bandgap silicon-containing copolymer, poly[2,7-(9,9dioctyl-silafluorene)-alt-5,5-(4,7-di-2-thienyl-2,1,3-benzothiadiazole)] (29).^[19] PCE of the PSC based on 29 as donor blended with PCBM (1:2 w/w) reached 5.4% (AM 1.5 G, $80 \,\mathrm{mW \, cm^{-2}}$) without additional post-annealing treatment and without any additive to control film morphology, which makes this class of materials among the best conjugated polymers reported up to now. The high efficiency is attributed to its lower HOMO level, broader absorption (350-750 nm) and higher hole mobility $(1 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$, FET method).^[19]

Compared to the fluorene unit, the carbazole moiety has good hole transporting property and a better chemical stability. A carbazole derivative, poly[*N*-9-heptadecanyl-2,7-carbazole*alt*-5,5-(4,7-di-2-thienyl-2,1,3-benzothiadiazole)] (**30**) with optical bandgap (1.88 eV) was reported by Leclerc and coworkers.^[20] Photovoltaic cells based on **30** as donor blended with PCBM (1:4 w/w) show a PCE of 3.6% (AM1.5G, 90 mW cm⁻²). The high efficiency is ascribed to the combination of broad absorption and good hole transporting ability, but the hole mobility is not reported.^[20]

A platinum metallopolyyne (31) with a low bandgap of 1.85 eV was recently reported by Wong et al.^[21] The PSC based on the blend of 31/PCBM (1:4, w/w) showed an average PCE of 4.1% without annealing. It is noteworthy that the device exhibits very high $J_{\rm sc}$ of 13.1 mA cm⁻² and relatively high open circuit voltage $V_{\rm oc}$ of 0.82 V. This is the first time that a metallated polymer has been applied in a PSC to get a high efficiency. The results demonstrate that metallated conjugated polymers are promising for application in high-performance PSCs.

Although reducing the bandgap of the conjugated polymers leads to a red shift of their absorption spectra and better absorption spectral matching with the solar spectrum, which benefits the sunlight harvest, it usually results in higher HOMO or lower LUMO energy levels of the conjugated polymers. For the polymer donor, higher HOMO results in lower open circuit voltage of the PSCs, and lower LUMO may influence the exciton charge separation on the donor/acceptor interface, which will decrease the efficiency. In addition, low-bandgap polymers may lose their short wavelength absorption. Therefore, most of the low-bandgap polymers did not show high efficiency. One has to balance the bandgap, width of the absorption band, and the HOMO and LUMO energy levels in designing low-bandgap polymers. The lowest bandgap reported to date for the new conjugated polymer donors with PCE higher than 1% is 1.2 eV for a thieno[3,4-b]pyrazine-based polymer (32).^[22] The PCE of the PSC based on 32 as the donor blended with PCBM (1:4 w/w) is 1.1%.

Besides being able to be used as a donor, a low-bandgap polymer can also be used as an acceptor in a PSC if its LUMO energy level is low enough. An alternative copolymer (**33**) of perylene diimide (PDI) and dithienothiophene synthesized by Zhan et al.^[23] shows a low E_g of 1.46 eV, high electron affinity with a LUMO level of -3.9 eV (similar to the LUMO level of PCBM), and high electron mobility of $1.3 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.^[23] The copolymer exhibits broad absorption throughout the visible and into the near-IR region (see Fig. 2d). All PSCs were fabricated with the blend **33** as acceptor and **8** as donor (1:1 w/w); the devices showed a PCE over 1%.^[23]

5. Conclusions and Outlook

Up to now, great efforts have been devoted to low-bandgap polymers and broad-absorption polymers with conjugated side chains for high-performance photovoltaic materials. The copolymerized D-A structure has been successfully used in obtaining low-bandgap polymers, especially copolymers containing fluorene or fluorene-like units (replacement of two benzene rings by two thiophene rings, or replacement of 9-C by Si or N) show high photovoltaic efficiencies with a maximum value of 5.5%. Broad absorption was realized by attaching conjugated side chains to the main chains of polythiophene and poly(thienylenevinylene) or by copolymerized D-A structure. The PT derivative with bi(thienylenevinylene) conjugated side chains displays strong and broad absorption from 350 nm to 650 nm, and a PCE of 3.18% was achieved for the PSC based on the polymer as donor. Higher hole mobilities on the order of 10^{-3} cm² V⁻¹ s⁻¹ were achieved for side-chain-conjugated polymers, crosslinked PTs with conjugated bridges, and lowbandgap polymers.

For the application in bulk-heterojunction-type PSCs, the preferred properties of the conjugated polymers include good solubility for solution-processing and film-forming ability, a strong and broad absorption band in the vis-NIR region for good sunlight harvesting, and high purity and high charge carrier mobility for high charge transport efficiency. In addition, the matching of the LUMO and HOMO energy levels of the donor and acceptor materials in PSCs is also very important for efficient charge separation and high open circuit voltage. For further improvement of the photovoltaic properties of the conjugated polymers, efforts should be devoted to the precise control of their HOMO and LUMO energy levels, structural modification for forming a preferred morphology in the blend film with PCBM, strong and broad visible absorption with high charge carrier mobility, high purity, and structural regularity. The commercial application of PSCs becomes more likely with the development of high efficiency conjugated polymer photovoltaic materials.

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