Russian Chemical Reviews **83** (7) 575 – 599 (2014) © 2014 Russian Academy of Sciences and Turpion Ltd

DOI 10.1070/RC2014v083n07ABEH004417

Organic bulk heterojunction photovoltaic structures: design, morphology and properties

G V Bulavko,[a] A A Ishchenko [b]

[a] *Institute of High Technologies, Taras Shevchenko National University of Kyiv prosp. Hlushkov 4G, 02033 Kyiv, Ukraine*
[b] *Institute of Organic Chemistry, National Academy of Sciences of Ukraine ul. Murmanska 5, 02660 Kyiv, Ukraine*

Main approaches to the design of organic bulk heterojunction photovoltaic structures are generalized and systematized. Novel photovoltaic materials based on fullerenes, organic dyes and related compounds, graphene, conjugated polymers and dendrimers are considered. The emphasis is placed on correlations between the chemical structure and properties of materials. The effect of morphology of the photoactive layer on the photovoltaic properties of devices is analyzed. Main methods of optimization of the photovoltaic properties are outlined.
The bibliography includes 338 references.

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I. Introduction

In 1953, researchers from Bell Labs† (New Jersey, USA) discovered the generation of electric current upon exposure of contaminated silicon pieces to sun light. This phenomenon is basic to the operation of the first silicon solar cell invented at the same research centre and to the design of photovoltaic materials.[1, 2]

Organic photovoltaic materials have been produced since the mid-1980s. Photovoltaic devices made of them are cheaper and much simpler in production than inorganic analogues. The ability of organic photovoltaic materials to form nanocomposites makes it possible to fabricate the active layer with a desired morphology and better adaptability to the shape and dimensions of photovoltaic devices.[3] A no less important point is the relative ease of variation of the structure and chemical composition of polymers in order to optimize their properties in targeted manner depending on the particular application of the final material.[4] This makes it possible to fabricate large thin-film, flexible panels based on organic compounds that are necessary to solve some problems in solar power engineering.[5]

The general principle of operation of an organic photovoltaic device is as follows. A mechanical blend or covalently bonded donor and acceptor components are placed between two electrodes with different work functions. Illumination of the system causes a transition of an electron from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO) of the donor or acceptor depending on the absorption band to which excitation occurs. Excitation of the donor component is followed by nonradiative electron transfer to the acceptor LUMO. Absorption of light by the acceptor component is followed by a similar electron transfer from the donor HOMO to the lowest singly occupied molecular orbital of

G V Bulavko post-graduate student of the Department of Supramolecular Chemistry and Biochemistry at the IHT, Taras Shevchenko National University of Kyiv.
Telephone: (380-44) 521 35 66, e-mail: gennadiybulavko@mail.ru
Current research interests: electronic processes in organic photosemiconductors.
A A Ishchenko Doctor of Chemical Sciences, Professor, Head of the Colour and Structure of Organic Compounds Department at the IOC NAS of Ukraine.
Telephone: (380-44) 499 46 04, e-mail: alexish@i.com.ua
Current research interests: electronic structure and photonics of organic molecules, light-sensitive materials.

Received 2 July 2013
Uspekhi Khimii **83** (7) 575 – 599 (2014); translated by A M Raevskiy

† Previously known as 'AT&T Bell Laboratories' and 'Bell Telephone Laboratories'.

acceptor. As a result, a positively charged centre (hole) appears on the donor and a negatively charged centre (electron) appears on the acceptor. The potential difference between electrodes predetermines transport of charge carriers, namely, electrons move towards the electrode with the lower work function (cathode) while holes move towards the electrode with the higher work function (anode), thus generating an electric current.[4,6]

In the first photovoltaic devices, the charge separation was achieved involving a potential barrier in the metal/semiconductor contact layer (Schottky barrier).[7–9] However, such devices are inefficient since photogeneration of charge carriers in them occurs only within a thin layer near the metal–organic layer interface. The photocurrent also decreases due to quenching of excitons at this interface.[10]

In the best organic photovoltaic structures, charge carriers are generated within a boundary layer between two different organic semiconductors (heterojunction structures).[1] Such structures can be made of different materials using diverse methods (by solution cast of polymeric composites with subsequent evaporation of solvent[1] or by vacuum deposition). Two types of photovoltaic structures are utilized, *viz*., layered and bulk heterojunction systems.

In layered structures, the donor and acceptor molecules having n-type and p-type conductivity, respectively, are localized in different layers.[2] Photogeneration of charge carriers in them occurs at the boundary separating two layers (a heterojunction).[1] The main advantage of layered structures consists in the absence of the main side process, *viz*., recombination of photogenerated charge carriers (holes and electrons) through separation of photoactive layers.[11] However, it is to a great extent leveled since photogeneration of charge carriers occurs only within a thin layer near the heterojunction, but this region absorbs only a small fraction of incident light which thus limits the quantum efficiency of the device.

In bulk heterojunction photovoltaic structures (BHPS),[2] both donor and acceptor fragments are located within the same photoactive layer. Usually, a film fabricated from organic compounds with n- and p-type conductivity is placed between two electrodes with different work functions.[12] Diverse contacts between the donor and acceptor molecules in the bulk of the active layer ensure a rather high concentration of photogenerated charge carriers. At present, BHPS exhibit the highest efficiency and this review is concerned with just the BHPS.

The scope of numerous reviews on organic photovoltaics reported to date is rather wide and includes:

— various types of photovoltaic cells with layer-by-layer deposition, BHPS, tandem ones, as well as main methods for improvement of their efficiency;[11,13]

— photonic properties of conjugated organometallic polymers;[14]

— photovoltaic devices based on polymers[15–17] and fullerene–polymer systems[18] and methods for improvement of their performance;

— application of conjugated polyelectrolytes in organic photovoltaics (a Section in Ref. 19)

— analysis of mainly physical processes in photovoltaic cells and construction of relevant physical models and mathematical formalism to describe them;[20]

— brief outline of main types of photovoltaic devices;[21,22]

— photovoltaic devices based on various polymer–nanoparticle systems;[23]

— detailed consideration of diverse types of organic photovoltaic structures;[24]

— the future of photovoltaics as branch of science (last Section in Ref. 25);

— effect of doping on the properties of organic photovoltaic devices;[26]

— analysis and generalization of experimental data on application of colloidal quantum dots in solar cells, as well as various types, design and operating principles of the solar cells.[27]

A particular place is occupied by the publications[13,28–31] concerning the design and application of novel graphene-based materials in photovoltaic devices.

Of considerable interest are dye-sensitized solar cells (DSSC).[32] The choice of dyes for the DSSC has been studied[33–35] and methods for improvement of their performance were also analyzed.[36–40] Such photovoltaic devices can not be treated as pure organic ones; they have been described elsewhere[41–44] and we will leave them out of consideration.

There is considerable literature on organic dyes in photovoltaic devices. However, the results of corresponding studies were not generalized in the reviews mentioned above. Also, those reviews provided no relevant information on recent advances in the application of conjugated polymers in photovoltaics. Graphene and its derivatives were studied only in connection with application in DSSC and polymeric photovoltaic devices. Also, little attention has been paid to the influence of morphology of the photoactive layer on the properties of BHPS, although the results of many studies show that the morphology to a great extent determines the efficiency of photovoltaic devices. In this review, we systematized and analyzed modern approaches to the design of efficient BHPS. To this end, we will also consider promising novel materials based on organic dyes and related compounds, conjugated polymers, dendrimers and graphene. The role of morphology of the photoactive layer in BHPS will be revealed.

II. Main parameters of organic photovoltaic devices

Photovoltaic devices are characterized by the short-circuit current (I_{sc}), the open-circuit voltage (V_{oc}) and the fill factor (FF)

$$\mathrm{FF} = \frac{P_{max}}{P_p} = \frac{I_{max}\,V_{max}}{I_{sc}\,V_{oc}} \qquad (1)$$

where P_{max} is the maximum power, P_p is the potential power and I_{max} and V_{max} are respectively the electric current and voltage at maximum actual power of the device.[1]

The efficiency of energy conversion by a device (η)‡ is given by the ratio[1]

$$\eta = \frac{P_{max}}{P_o} \qquad (2)$$

where P_o is the optical power.

‡ This parameter characterizes the performance of the photovoltaic device.

The spectral response of an organic photovoltaic device is an important parameter which can be used for optimization of the operation of such devices. The photocurrent is measured as a function of the wavelength of incident light and then is compared with the illumination intensity. The efficiency of conversion of incident photons to electricity is characterized by the external quantum efficiency (EQE) which is equal to the number of knocked-out electrons per incident photon,

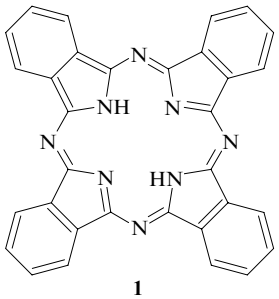
$$\mathrm{EQE} = \frac{N_e}{N_{ph}} = \frac{I_{sc}\, h\, c}{P_o\, \lambda\, e} \qquad (3)$$

where N_e is the number of knocked-out electrons, N_{ph} is the number of incident photons, h is the Planck constant, c is speed of light in vacuum, λ is the wavelength of incident light and e is the electron charge.

III. Photovoltaic structures containing organic dyes and related compounds

The first bilayer organic photovoltaic structures were fabricated by mixing dyes that could provide electron transfer (rhodamines or triphenylmethanes) with dyes that could provide hole transport (phthalocyanines, *e.g.*, tetraazabenzoporphyrin (**1**), or merocyanines).[45]

Structure 1

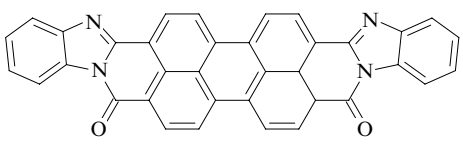


1

Such BHPS can generate a voltage to 200 mV and a photocurrent of about 10^{-8} A at low illumination intensity. In 1984, Harima *et al.*[46] reported efficient photogeneration of charge carriers at the heterojunction between Zn phthalocyanine (**2a**) and 5,10,15,20-tetrakis(3-pyridyl)porphyrin (**3**).

Tang[47] reported a nearly 1% energy conversion efficiency for a photovoltaic cell under simulated sun light source. The structure was fabricated by deposition of a Cu-phthalocyanine (**2b**) layer on a glassy substrate preliminarily coated with indium tin oxide (ITO) and then with a layer of 3,4,9,10-perylenetetracarboxylic bisbenzimidazole (PTCBI) (**4**). A silver cathode was utilized as the top layer of the device.

Structures 2–4



4

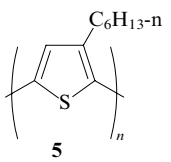
At an illumination intensity of 75 mW cm^{-2} (this corresponds to sun light at a Sun angle of 60.2°), the energy conversion efficiency was ~0.95%.

Bulk heterojunction photovoltaic structures based on polymer–fullerene blends have been intensively developed since the 1990s.[48–52] Such blends are relatively cheap, have good physicomechanical properties (light weight, flexibility) and exhibit a rather high energy conversion efficiency. This is due to the fact that fullerene is a strong electron acceptor whose molecule has the shape of a sphere, which makes recombination of photogenerated charge carriers difficult compared to the case of planar acceptor molecules with comparable electron-withdrawing ability (*e.g.*, tetranitrofluorenone).[53]

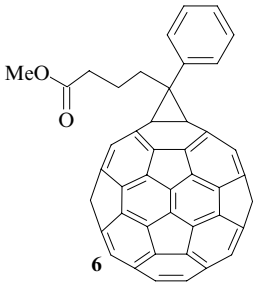
Some of the most efficient organic BHPS were prepared using fullerene derivative/conjugated polymer blends.[54, 55]

Currently, the most widely used BHPS contain poly(3-n-hexylthiophene) (P3HT) (**5**) as a donor and [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) (**6**) as an acceptor.[49]

Structures 5, 6



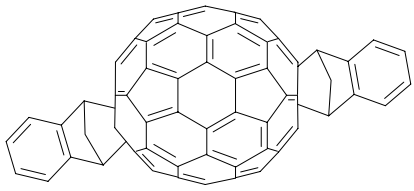
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6

The energy conversion efficiency values reached by the organic photovoltaic devices with the active layer made of P3HT/fullerene derivative blends were ~5% for the P3HT/PCBM system[56] and 6.69% for the P3HT/C_{70} fullerene derivative (**7**) system.[57] Theoretically, the efficiency of polymer/fullerene derivative blends can exceed 10% for single-layer and 15% for bilayer photovoltaic cells.[58, 59] An energy conversion efficiency of >7% was reported for polymeric photovoltaic cells.[17]

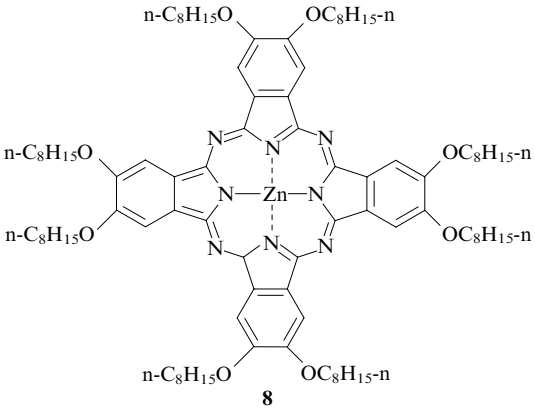
Structure 7



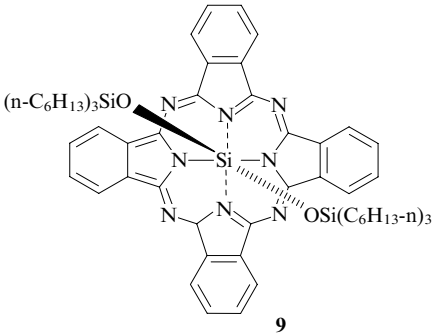
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However, P3HT can absorb only ~25% of the total number of incident photons. Naturally, it was necessary to design systems with higher light absorption. A suitable method involves doping the photoactive layer with an organic dye that absorbs in a longer-wavelength region of the sun light spectrum where the main components of the photovoltaic cell do not absorb. However, introduction of the dye may appear to be inefficient or even reduce the performance of the device. The latter occurs if the dye forms aggregates, which reduces the efficiency of light absorption or the mobility of charge carriers in the active layer.[60–62] For instance, structures based on P3HT/PCBM were doped with dyes absorbing in the near-IR region, *viz.*, derivatives of Zn-phthalocyanine **8** and Si-phthalocyanine **9**.[48]

Structures 8, 9



8



9

The absorption spectra of the systems P3HT/PCBM/**8** and P3HT/PCBM/**9** in the form of solutions and films were compared.[48] The absorption spectra of the solutions represented superpositions of the spectra of P3HT and corresponding dyes and thus indicated uniform mixing, no decomposition to components and no interactions between the components in the ground state. The absorption spectrum of the P3HT/PCBM/**8** films was quite different, namely, the absorption band of dye **8** at about 670 nm was rather broad and weak compared to the corresponding band in the spectrum of the solution. This indicated the formation of aggregates of compound **8** and corresponding decrease in the effective absorption coefficient of the films.[63,64] The spectra of the P3HT/PCBM/**9** films showed no such changes. The photovoltaic parameters of the devices measured before and after annealing of the films are listed in Table 1.

The $I-V$ curve of the structure P3HT/PCBM/**8** measured before thermal annealing almost matched that of the dye-free reference sample. After thermal annealing, the I_{sc} values of the device and the reference sample considerably increased, whereas the FF value of the device appeared to be much lower than that of the reference sample. Doping with dye **8** caused the η value to be halved (see Table 1); this was explained by aggregation of the dye in the BHPS films and thus limitation of transfer of photogenerated charges at the interface.[65,66] Contrary to these results, the η value for the P3HT/PCBM/**9** system increased to 2.7% while the FF value remained equal to that of the reference sample.

Differences in the $I-V$ curves of the systems under study are due to different ability of the dyes to undergo aggregation,[48] namely, stacking of the molecules of dye **8** along the normal to the phthalocyanine plane leads to formation of sandwich-type aggregates; the molecule of dye **9** contains

Table 1. Parameters of BHPS based on P3HT/PCBM systems containing dyes **8** and **9**.[48]

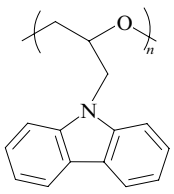
Cell	I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
Before thermal annealing				
P3HT/PCBM [a]	1.5	0.6	33	0.3
P3HT/PCBM/**8**	1.9	0.58	32	0.35
P3HT/PCBM/**9**	2.1	0.64	43	0.58
After thermal annealing				
P3HT/PCBM [a]	6.5	0.58	59	2.2
P3HT/PCBM/**8**	6.2	0.48	37	1.1
P3HT/PCBM/**9**	7.9	0.58	59	2.7

[a] Reference sample.

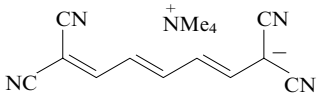
two bulky groups in the axial direction perpendicular to the phthalocyanine plane, which precludes the formation of this type of aggregates. Taking into account this fact, considerable attention should be paid to the development of methods for optimization of the dye structures and the properties of the medium as steps in the design of BHPS. Relevant methods were reviewed elsewhere.[67,68]

However, in some cases aggregation may have a positive effect. In particular, it was traditionally accepted that photoconductivity may appear only if a donor and an acceptor are utilized.[69] However, an unusual effect, namely, photoconductivity in highly concentrated blends of two electron donors, the oligomer poly(*N*-epoxypropylcarbazole) (PEPC) **10** and anionic polymethine dye (An5)$^-$ **11**, was reported recently.

Structures 10, 11



10



11

The effect is due to the formation of associates of contact ion pairs of the dye, in which intramolecular electron transfer resulting in a neutral radical of the dye (An5)$^\bullet$ occurred. According to quantum chemical calculations, the formation of (An5)$^\bullet$ is accompanied by a decrease in its HOMO energy that becomes lower than that of *N*-methylcarbazole, a PEPC unit (Fig. 1); therefore, (An5)$^\bullet$ acts as an efficient electron acceptor. As the radical captures an electron from the HOMO of the carbazole nucleus, a radical cation, *i.e.*, a hole appears. Thus, mixing of two donors taken in high concentrations causes the onset of p-type conduction.[70]

Mutual arrangement of molecules of the third component and molecules of other components in the films prepared from BHPS blends plays an important role.

Charge transfer in BHPS with the P3HT/**9**/PCBM active layer is schematically shown in Fig. 2 *a*. As can be seen, the molecule of dye **9** located near the P3HT/PCBM heterojunction can inject electrons into PCBM and holes into P3HT because its energy levels lie in the LUMO region of P3HT and in the HOMO region of PCBM.[71–74] If mole-

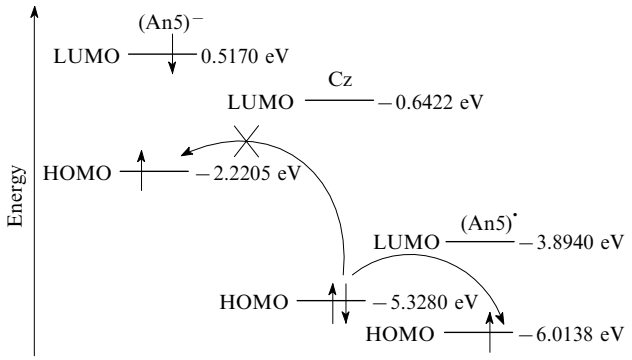


Figure 1. Energy levels and possible photoinduced electron transitions in carbazole (Cz), anionic dye $(An5)^-$ and its neutral radical $(An5)^{\bullet}$.[69]
Energies were obtained from B3LYP/6–31G(d,p) density functional quantum chemical calculations.

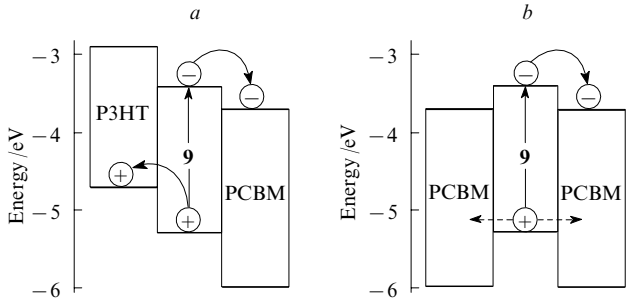


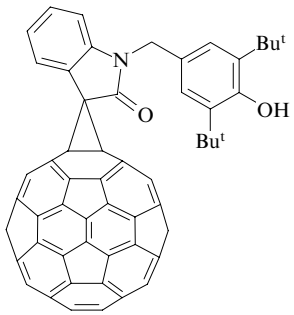
Figure 2. Charge transfer in a BHPS with the P3HT/**9**/PCBM active layer and the HOMO/LUMO energy diagrams for the molecules of P3HT, PCBM and compound **9**.[48]
(*a*) Dye **9** is located at the boundary between P3HT and PCBM; (*b*) dye **9** is localized within the PCBM domain.

cules **9** are distributed within the PCBM layer as shown in Fig. 2 *b*, excitons of this compound can inject electrons into the PCBM layer but injection of holes into PCBM domains is impossible because the HOMO level of PCBM has a lower energy. This also holds for excitons of compound **9** confined within the P3HT domains. Therefore, only the molecules of dye **9** located at the P3HT/PCBM interface can contribute to the photocurrent. After thermal annealing, the number of such molecules increases. However, a number of dye molecules are at the P3HT/PCBM interface before thermal annealing.[75, 76]

The action mechanism of the dye in P3HT/PCBM/**9** systems was studied.[48] There are two aspects. One of them is related to additional absorption of photons upon introduction of the dye **9**; this favours an increase in I_{sc}. In our case, this aspect makes a small contribution owing to a rather narrow absorption band of compound **9**. The other aspect is due to the fact that molecules **9** do not directly absorb light and thus favour charge separation in the P3HT excitons within the interface. Thus, the dye molecules favour energy transfer from the donor to the acceptor.

An important problem in the design of BHPS is the choice of the acceptor. Researchers from the Russian Federation synthesized a new fullerene-containing acceptor **12** for organic photovoltaic devices.[77]

Structure 12



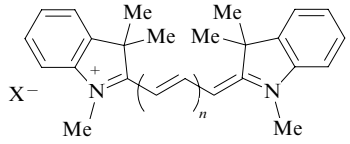
12

Compound **12** is characterized by more intense absorption in the wavelength range from 250 to 600 nm compared to PCBM; this leads to better absorption of light by the photovoltaic device. However, acceptors based on C_{60} fullerene and its derivatives do not absorb light with wavelengths > 600 nm; therefore, nearly 60% photons are not absorbed.[78–81] The role of acceptors in BHPS can be successfully played by polymethine dyes.[78, 82–86] They are characterized by the greatest absorption range and by record high extinction coefficients among organic dyes.[87, 88] Utilization of polymethine dyes allows one not only to span the entire visible region (dyes **13**–**16**),[82, 84] but also to extend the IR spectral sensitivity from 850 nm (dyes **15** and **17a**,**b**) [78, 82] to 1400 nm (dye **18**).[85, 86] Possible donors include PEPC **10** and its derivatives for the dyes **13a**–**c**, **15** and **18** [82, 84–86] and poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) **19** for the dyes **14a**,**b**, **16** and **17a**,**b**.[78, 83]

Studies of the Stern–Volmer concentration dependences of luminescence quenching and instantaneous absorption spectra in the case of flash photolysis revealed that the photovoltaic effect in blends of polymers (PEPC and MEH-PPV) with cationic polymethine dyes occurs through electron transfer from the polymer to the dye molecule upon photoexcitation of the latter.[78, 82] The results of experimental determination of the redox potentials by cyclic voltammetry and the results of quantum chemical calculations of the HOMO and LUMO energies of polymethines and polymers showed that the photovoltaic effect is due to appropriate mutual position of the frontier orbitals of polymethines and those of polymers. It is noteworthy that the photovoltaic effect in polymethines can be smoothly controlled by varying the key structural parameters, namely, the length of the polymethine chain (*n* value) and the electron-donating ability of terminal groups. For instance, the photovoltaic effect in the series of dyes **13c**, **14c**, **15** becomes regularly less pronounced because the band gap decreases with increasing *n* mainly owing to an increase in the HOMO energy of the dye.[84]

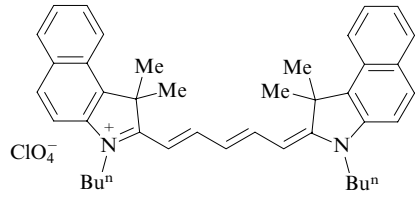
Note that variations of less important structural fragments can also significantly influence the output parameters of the BHPS based on polymethine dyes. For instance, replacement of ethyl substituent (dye **17a**) by octadecyl one (dye **17b**) causes the photocurrent to reduce by more than eight times.[78] This was explained by the fact that the long-chain octadecyl substituent acts as a spacer between the donor and the acceptor and reduces the electronic

Structures 13–19

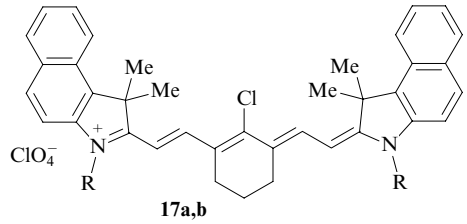


13–15

n = 1: X = Cl (**13a**), I (**13b**), BF_4 (**13c**); n = 2: X = PF_6 (**14a**), Cl (**14b**), BF_4 (**14c**); n = 3: X = BF_4 (**15**)

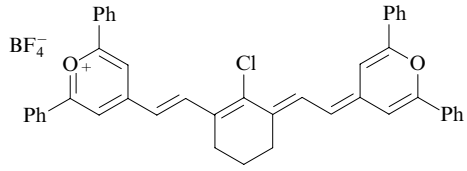


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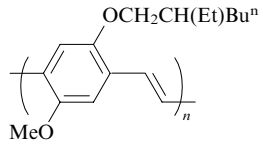


17a,b

R = Et (**a**), n-$C_{18}H_{37}$ (**b**)



18



19

coupling and thus the photoinduced charge transfer between them.

It was traditionally assumed that the conductivity of polymer semiconductors doped with ionic dyes is governed by the structure of the ionic chromophore, being independent of the nature of the colourless counterion.[4,89] Taking dyes **13a–c** in PEPC as examples, it was found that the nature of the counterion can play an important role in the photogeneration and relaxation of electron-hole pairs.[90] This is due to the fact that salt-like dyes in polymeric matrices exist as solvent-separated and contact ion pairs.[87,88] In the latter case, the counterion may have a strong effect on the spectral-luminescent properties,[87,88] internal conversion and intersystem crossing.[87,88] For instance, replacement of chloride anion (dye **13a**) by iodide anion (dye **13b**) led to enhanced intersystem crossing owing to the effect of the heavy atom, to an increase in the concentration of long-lived triplet excitons and, as a consequence, to a considerable increase in the photocurrent.[90] The role of triplet excitons in polymethine dyes incorporated into PEPC has been studied in detail from luminescence quenching in external magnetic field.[90–92]

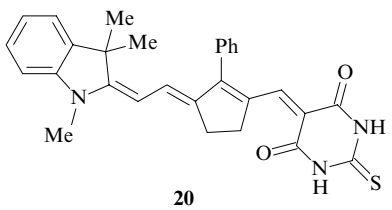
An interesting effect of the counterion in the photovoltaic structures based on the systems C_{60}/**14a** and C_{60}/**14b** was observed.[83,93] Being a typical electron acceptor, C_{60} fullerene behaves conventionally in structure C_{60}/**14a**, but acts as an electron donor in structure C_{60}/**14b**. Such a reversal of the donor-acceptor properties can be detected from the changes in the direction of the photocurrent. This was explained by different mobilities of PF_6^- and Cl^- anions of dyes **14a** and **14b**, respectively.[83] Anions diffuse towards C_{60} fullerene molecules and produce an additional electric field in its vicinity. The ion redistribution time is ~100 min for dye **14a** and 10 min for compound **14b**. It is believed that positions of the energy levels of the dye and fullerene and, consequently, their donor-acceptor properties change under the action of Cl^- anion that more efficiently penetrates the C_{60} layer. As a result, C_{60} in the C_{60}/**14b** system becomes an electron donor while dye **14b** becomes an electron acceptor.

We believe this is not a rigorous explanation because the anions, PF_6^- and Cl^-, differ appreciably not only in size, but also in redox properties; namely, Cl^- having a much lower oxidation potential[94] can, unlike PF_6^-, be involved in photoinduced electron transfer in ion pairs of cationic dyes.[94–96] The process is also facilitated since dye **14a** forms more tightly bound pairs than compound **14b**.[97] As a result of photoinduced electron transfer, the dye anion is transformed to a neutral radical that may appear to be capable of withdrawing an electron from C_{60} fullerene, thus turning it into an electron donor. To draw unambiguous conclusions on the mechanism of the photovoltaic effect,[83] one should synthesize cationic polymethine dyes with various anions and study their photophysical and electrophysical properties. For comparison, it seems appropriate to have pairs of anions with similar ionic radii and strongly different redox potentials and *vice versa*. These studies should make the search for optimal dyes for BHPS much easier.

If adverse effects of counterions are expected, intraionic dyes (in particular, merocyanines) should be used instead of ionic dyes.[98] At first glance, this may appear to be of little promise because merocyanines usually absorb in the visible region and have lower extinction coefficients than corresponding ionic dyes.[98] However, the possibility to synthesize merocyanines with spectral parameters similar to those of corresponding ionic dyes was reported.[99] Namely, merocyanine **20** is characterized by strong near-IR absorption comparable with that of cationic dye **15** at nearly the same position of the absorption maximum.

These properties of merocyanine **20** are due to thorough 'tuning' of the donor-acceptor properties of terminal heterocyclic groups, the length of the polymethine chain

Structure 20



20

and the introduction of a cyclic fragment.[99] These structural factors allowed the electronic structure of merocyanine **20** to approach that of a ideal polymethine characterized by the maximum degree of equalization of the bond orders and by alternation of positive and negative charges in the chromophore.[98] Merocyanine **20** had better photovoltaic parameters than cationic dye **15** due to the absence of the side dark effects in the former (these effects in system **15** were caused by the counterion).[99] The design of novel merocyanines with the structure of a ideal polymethine, capable of absorbing light in the near-IR region seems to be promising for further progress in organic photovoltaics.

Yet another study on the photovoltaic properties of structures based on polymethine dyes is available.[84] Samples were prepared in the form of free-surface structures, namely, an ITO layer was coated with composites comprising a PEPC analogue and dyes **13c**, **14c** and **15**. Unlike sandwich structures, free-surface structures do not experience the effect of contact potential difference which influences the dissociation of excitons and transport of charge carriers. The efficiency of production of photogenerated charge carrier in the free-surface structures can be evaluated by measuring the surface potential of films with a dynamic probe (Kelvin technique).[100] The photo-emf was 415, 160 and 120 mV for the structures based on dyes **13c**, **14c** and **15**, respectively. The advantages of the free-surface structures include simple fabrication procedure and technological processing as well as the possibility of rapid evaluation and effective investigation of photogeneration and charge carrier transportation. Studies of these processes represent a key step of research in the field of photovoltaics. Their results can be used in the future design of novel BHPS employing materials that were not utilized previously.

Bulk heterojunction photovoltaic structures can be based not only on typical dyes, but also on a large number of related organic chromophores, usually, low-molecular-mass conjugated systems exhibiting the properties of semiconductors.

The introduction of 3,6-dimethyl-9,10-diphenylphenanthrene (**21**) as a material for hole transport into a photovoltaic device based on P3HT/PCBM causes the energy conversion efficiency and the short-circuit current to increase by 16% and 25%, respectively.[101]

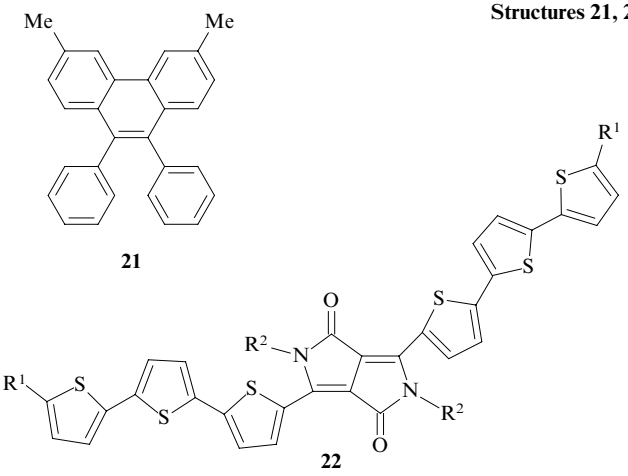
Bulk heterojunction photovoltaic structures based on P3HT/$PC_{70}BM$ blends containing different amounts of organic semiconductor **22** absorbing light in the near-IR region were studied.[49]

The case in point is that P3HT is not a good electron donor. Different authors estimate its band gap (E_g) as follows:

E_g/eV	1.9–2	1.7–2.1	1.9
Ref.	102	103	57

To improve the performance, such structures are doped with electron donors, *viz.*, molecules of low-molecular-mass compounds.[60, 104] When choosing the donor molecules, it is important that their band-gap levels lie between the energy levels of the polymer and fullerene. When compound **22** was used as an electron donor for the system P3HT/$PC_{70}BM$, the I_{sc} value increased from 7.7 to 8.6 mA cm^{-2} and V_{oc} increased from 0.60 to 0.63 V relative to those of the reference device containing no compound **22**. The effect is achieved as a result of extension of the absorption range,

Structures 21, 22

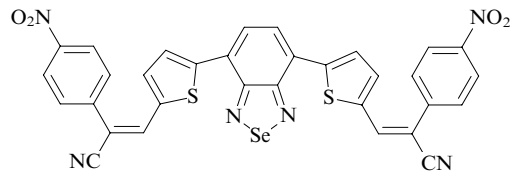


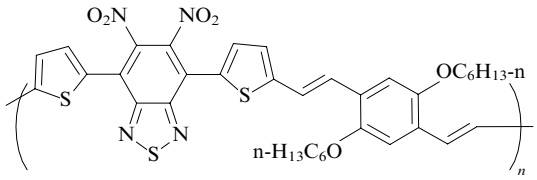
R^1 = n-C_6H_{13}, R^2 = $CH_2CH(Et)Bu^n$

which leads to an increase in the number of photogenerated charge carriers.[49]

Photovoltaic devices comprised of small molecules based on 2,1,3-benzoselenodiazole derivative **23** incorporated into a system based on a copolymer of **24** and PCBM were reported.[105]

Structures 23, 24





In the first step of research on the photovoltaic parameters, structures with the **23**/PCBM and **24**/PCBM active layers were prepared. Parameters of the first device appeared to be better (Table 2). However, in both cases the energy conversion efficiency was low. To make the process more efficient, the photoactive layer was prepared from blends of **24**/PCBM with compound **23** because the energy levels of molecules **23** lie between those of compound **24** and PCBM.[106, 107] In this case, improved performance of the photovoltaic device (see Table 2) can be explained by additional absorption in the long-wavelength region compared to that of copolymer **24**. Also, 2,1,3-benzoselenodiazole behaves as a donor responsible for the increase in the number of donor–acceptor interfaces and, consequently, photoinduced charge separation increases. The 2,1,3-benzoselenodiazole molecules can inject electrons into PCBM and holes into the copolymer only if they are at the interface; otherwise, they do not favour the appearance of photocurrent.

Table 2. Parameters of BHPS with different compositions of the photoactive layer.[105]

Photoactive layer	I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
24/PCBM	1.3	0.93	43	0.52
23/PCBM	2.7	0.98	49	1.3
24/PCBM/**23**	4.1	1.05	52	2.24
23/PCBM/**24** [a]	4.6	0.92	54	3.16

[a] Thermally annealed sample.

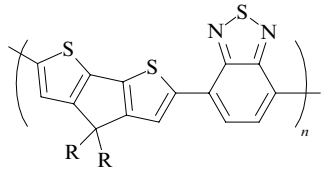
Different amounts of the low-band-gap conjugated polymer poly[2,6-(4,4-bis(2-ethyhexyl)-4*H*-cyclopenta[2,1-*b*;3,4-*b*′]-dithiophene-*alt*-4,7-(2,1,3-benzothiadiazole)] (PCPDTBT) (**25**) were added to a device based on P3HT/PCBM blend.[50]

The reasons to choose this polymer are as follows:

— the polymer absorbs in the near-IR region, which extends the absorption range of the photovoltaic cell;[108, 109]

— appropriate arrangement of the HOMO and LUMO of the polymer relative to the electron energy levels of P3HT and PCBM (Fig. 3), namely, the HOMO of polymer **25** lies between the HOMOs of P3HT and PCBM HOMOs; the same holds for the LUMOs. As a result, photoinduced charge transfer between P3HT and PCBM, between polymer **25** and PCBM, as well as between P3HT and **25** is energetically allowed.

Structure 25



25

R = $CH_2CH(Et)Bu^n$

With this design of the BHPS cell, both polymers favour generation of charge carriers similarly to the tandem structures that represent a number of stacked photovoltaic cells connected in series and separated by some heterojunctions.[110] However, the theoretical maximum of efficiency for three-component cells is lower than for the tandem ones.[111] Once compound **25** is added, the absolute efficiency increases, the EQE value decreases in the absorption range from 400 to 600 nm but increases in the near-IR region.

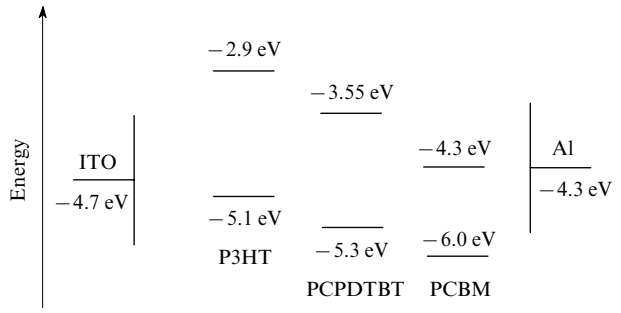
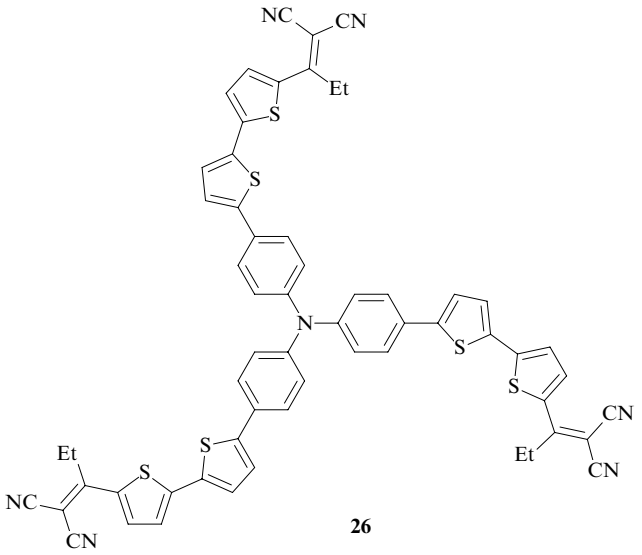


Figure 3. Energy levels of electrodes and organic semiconductors used in the device proposed in Ref. 50.
Energies were determined by cyclic voltammetry.

A possible charge transfer mechanism underlying the increase in the efficiency of the photovoltaic device is as follows.[50] Initially, light absorbed by PCPDTBT is converted to charge carriers by efficient electron transfer to PCBM. In the second step, positively charged carriers in PCPDTBT are efficiently transferred to the P3HT phase through which they are subsequenctly delivered to the electrode with relatively low bimolecular recombination losses. In addition, photoinduced electron transfer from P3HT to PCPDTBT contributes to the overall photocurrent generation in the BHPS cell.

Efficient photovoltaic devices based on tris[4-(5′-(2,2-dicyano-1-ethylvinyl)-2,2′-bithiophen-5-yl)phenyl]amine (**26**) having strong absorption in the region from 450 to 580 nm and various C_{60} and C_{70} fullerene derivatives were reported.[112]

Structure 26



26

A series of novel electron-donor materials based on tetrakis(oligothienyl)silanes for BHPS were synthesized and studied.[113] A photovoltaic cell with the photoactive layer prepared from oligomer **27** and $PC_{70}BM$ was characterized by the following parameters:

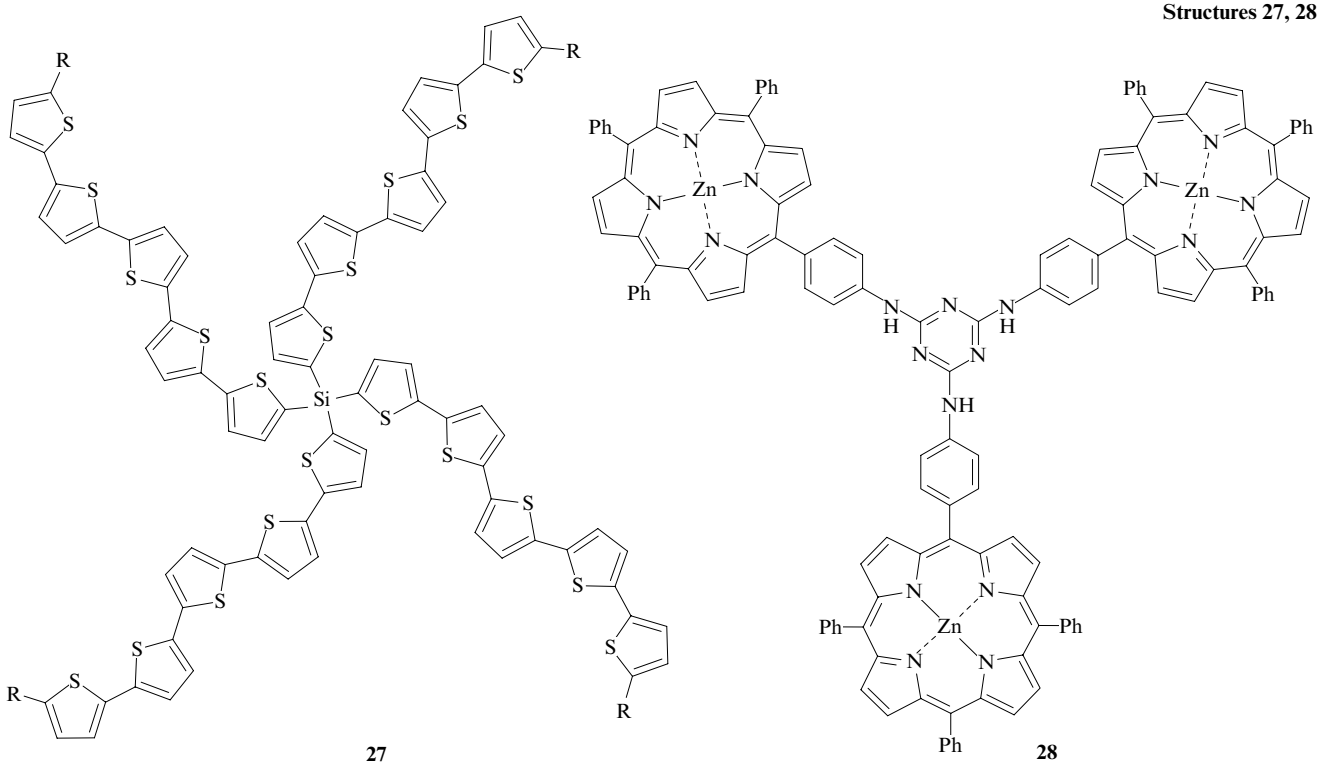
I_{sc}/mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
6	0.8	28	1.4

A BHPS based on a zinc complex of tris[4-(5,10,15-triphenylporphyrin-20-yl)phenylamino]-1,3,5-triazine (**28**) was reported.[114] The HOMO and LUMO energies of compound **28** are −5.5 eV and −3.5 eV, respectively; therefore, it can behave as a donor in blends with PCBM and as an acceptor in blends with P3HT.[114] The energy conversion efficiency was 0.5% in the former and about 0.1% in the latter case.

IV. Organic photovoltaic devices based on graphene derivatives

Graphene, which is characterized by high charge carrier mobility,[115, 116] good mechanical properties[117–119] and transparency,[120] immediately attracted the attention of

Structures 27, 28



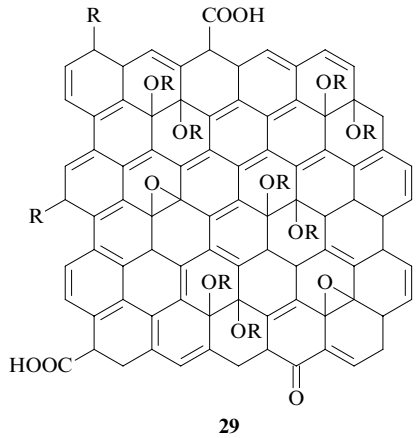
27 **28**

R = $CH_2CH(Et)Bu^n$

researchers who work in the field of organic photovoltaics.[121] The morphological and electrochemical properties of graphene derivatives were studied[122,123] and the possibility of their application as efficient electron-withdrawing materials was considered.[124–129]

Fabrication of BHPS with the active layer containing P3HT and functionalized graphene **29** was reported.[130]

Structure 29



29

R = C(O)NHPh

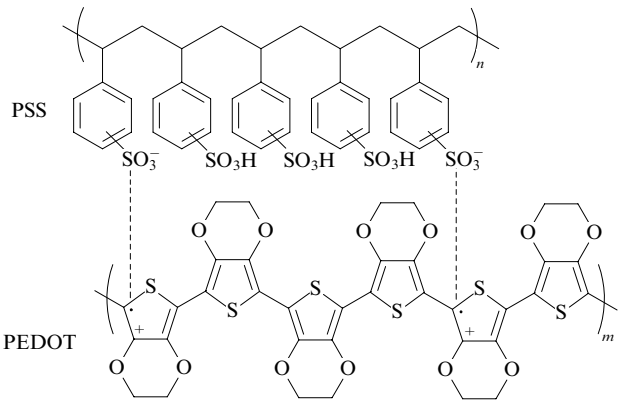
Functional groups were introduced to enhance the solubility of graphene. The content of the functionalized graphene was varied from 2.5 mass % to 15 mass %. To improve the performance, the photovoltaic device was thermally annealed. The best photovoltaic parameters were obtained for the device containing 10 mass % of functionalized graphene **29**:

I_{sc}/mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
3.3	0.88	39	1.1

Thermogravimetric analysis and differential scanning calorimetry studies showed that annealing is accompanied by removal of functional groups and reduction of π-conjugated domains in graphene; as a result, the charge carrier mobility significantly increases.[131]

Hole injection into photovoltaic cells is most often performed using a layer of poly(3,4-ethylenedioxythiophene)/polystyrenesulfonic acid (PEDOT : PSS) (**30**).

Structure 30



30

However, such a layer is deposited from highly acidic suspensions (pH 1), which may lead to corrosion of ITO at elevated temperatures.[132] Also, water from PEDOT : PSS may appear in the active layer; this will reduce the performance of the device.[133] It was proposed to use a thin layer of graphene oxide deposited from neutral aqueous suspensions instead of PEDOT : PSS.[134] Graphene oxide bears oxygen-containing functional groups; most often, these are epoxy and hydroxyl groups.[135–137] This leads to violation of the sp^2 hybridization of the hexagonal graphene sheet and the material becomes an insulator. The electronic structure of graphene oxide is nonuniform because of the presence of a mixture of the sp^2 and sp^3 hybridizations.[138] Probably, lateral transport of charge carriers occurs by hopping from localized sp^2 states to the Fermi level.[139] The corresponding density of states can be increased upon removal of some amount of oxygen atoms by heat treatment or chemical treatment which facilitates charge-carrier transport.[140–142] In a study of a BHPS based on P3HT/PCBM with a graphene oxide layer instead of PEDOT : PSS,[134] the work function for graphene oxide thin films was determined by Kelvin scanning probe microscopy. The experimentally determined work function value appeared to be somewhat higher than the value reported[143] for untreated graphene, *viz.*, ~ 4.9 *vs.* 4.6 eV, respectively. Samples with different thickness of the graphene oxide layer were prepared. The data presented in Table 3 allow one to compare the photovoltaic parameters of the samples under study and those containing the PEDOT : PSS layer.

Table 3. Parameters of BHPS containing different-type buffer layers.[134]

Layer	I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
ITO [a]	9.84	0.45	41.5	1.8
PEDOT : PSS	11.15	0.58	56.9	3.6
Graphene oxide				
2 nm	11.40	0.57	54.3	3.5
4 nm	10.22	0.57	33.9	2.0
10 nm	7.84	0.59	18.8	0.9

[a] A reference cell with no buffer layer.

The optimal thickness of the graphene oxide layer appeared to be 2 nm.[134] In this case, photovoltaic cells are at least competitive with the solar cells containing the PEDOT : PSS layer. Further increase in the thickness of the graphene oxide film led to deterioration of the operating properties of the cell.

The cost of ITO-based electrodes continuously increases due to complexity of fabrication and limited availability of indium.[144] In this connection, a search for novel transparent materials for fabrication of electrodes for optoelectronic devices has been carried out. Graphene has good optical and electrical properties and therefore seems to be a promising material for transparent electrodes.[145, 146] Indeed, graphene-based materials were successfully used as transparent electrodes for polymeric BHPS.[147–149] A comparison of the photovoltaic characteristics of the BHPS based on P3HT/PCBM with ITO and graphene anodes was reported.[150] A scheme of the energy levels for the photovoltaic cell with a graphene anode is shown in Fig. 4.§

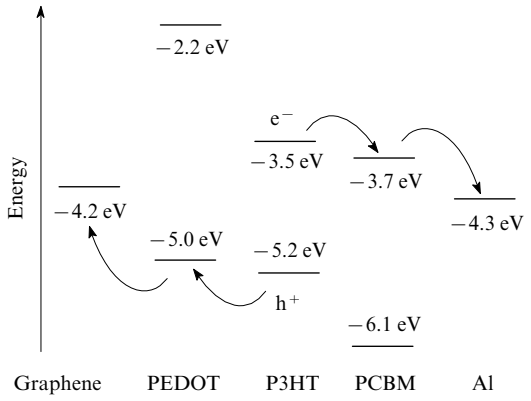


Figure 4. Energy levels and charge transport in BHPS with graphene anode: a schematic.[150]

The operating parameters of the structures with the graphene anode (Type I BHPS) and the ITO anode (Type II BHPS) are as follows:

Parameter	I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
BHPS I	1.18	0.46	24.58	0.13
BHPS II	12.32	0.6	48.57	3.59

It is believed that low energy conversion efficiency of Type I BHPS is due to hydrophobicity of the surface of the graphene film, which underlies a nonuniform distribution of the PEDOT : PSS layer. Taking into account this fact, it was assumed[150] that optimization of the graphene surface should lead to significant improvement of the performance of the cell. The use of graphene as a cathode material for organic photovoltaic devices was also reported.[151, 152]

The efficiency of the BHPS with graphene acceptors is much lower than that of the BHPS with fullerene acceptors, mainly due to the conducting properties of graphene, which considerably reduces the hole mobility and favours charge carrier recombination.[153] A method for doping photovoltaic structures based on P3HT/PCBM with graphene was proposed.[154] Graphite oxide prepared in the first step is then treated with phenyl isocyanate and sonicated to give modified graphene (SPF–Graphene)[155, 156] which can be dissolved in chlorobenzene and thus co-deposited with P3HT and PCBM. The photovoltaic characteristics of the BHPS containing different amounts of modified graphene are listed in Table 4.

It follows that devices containing 10 mass % of graphene exhibit the best properties; further increase in the graphene concentration leads to deterioration of properties, probably due to aggregation of graphene sheets and subsequent poorer charge transfer and enhanced charge carrier

§ It is our opinion that for the motion of charge carriers to occur as indicated in Fig. 4, the anode should be characterized by a higher work function than the Al cathode.

Table 4. Parameters of BHPS based on P3HT/PCBM doped with modified graphene.[154]

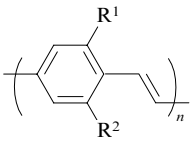
Graphene content (mass %)	I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
0	3.6	0.64	30	0.88
1	3.8	0.64	30	0.92
2.5	4.1	0.64	40	1.05
5	4.5	0.64	41	1.21
10	5.3	0.64	41	1.4
12.5	4.5	0.63	41	1.2
15	2.8	0.57	42	0.67

recombination.[157] The optimal morphology of the photoactive layer is achieved at a graphene content of 10 mass %. Doping the active layer with graphene offers additional channels of electron transfer from PCBM to cathode and less intense charge carrier recombination.[158] Graphene additives provide continuous electron transport and serve as a kind of 'conductive bridges' between PCBM and cathode.

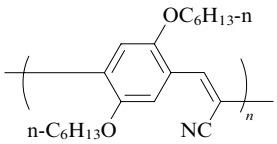
V. Novel conjugated polymers for photovoltaic structures

Conjugated polymers are widely used to produce photovoltaic devices. Devices based on, *e.g.*, poly(1,4-phenylenevinylene) (PPV) (**31a**), MEH-PPV, poly[2,5-bis(hexyloxy)-1,4-(1-cyanovinylene)phenylene] (CN-PPV) (**32**) and poly[2-methoxy-5-(3,7-dimethyloctyloxy)-1,4-phenylenevinylene] (MDMO-PPV) (**31b**) are characterized by relatively low energy conversion effciency ($\sim$2.5%).[159,160] Therefore, molecular design and synthesis of novel conjugated polymers with wide absorption ranges and low HOMO energies is topical. These can be, *e.g.*, conjugated donor-acceptor copolymers comprising donor and acceptor fragments. Low-band-gap donor–acceptor conjugated polymers, such as compounds **33** and **34**, can be used as the buffer layer in hybrid photovoltaic devices.[161,162]

Structures 31–34

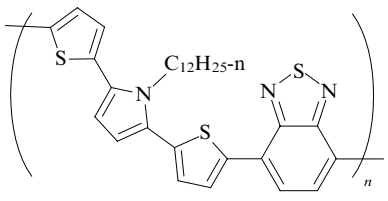


31a,b

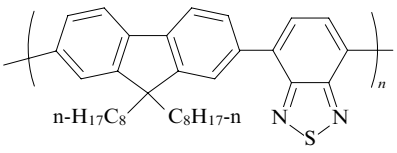


32

$R^1 = R^2 = H$ (**a**); $R^1 = O(CH_2)_2CH(Me)(CH_2)_3Pr^i$, $R^2 = OMe$ (**b**)



33



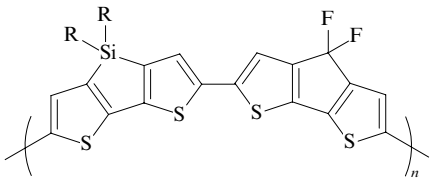
34

Low-bandgap conjugated polymers (E_g < 1.8 eV) as electron-donor materials for BHPS were reported.[163–165] The best results were obtained with the copolymers based on benzothiadiazole (*e.g.*, polymer **25**), namely, the energy conversion efficiency was in the range 1% to 5.5%.[105,111,166,167]

A series of cyclopentadithiophene-containing copolymers for photovoltaic cell applications was synthesized.[168] The highest energy conversion efficiency was achieved for a device based on copolymer **35** and PCBM.

I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
8.51	0.62	45	2.4

Structure 35



35

R = $CH_2CH(Et)Bu^n$

Yet another group of promising materials for organic photovoltaic devices includes conjugated polymers based on perylenediimide. In particular, photovoltaic cells based on blends of alternating conjugated copolymer **36** with P3HT were reported.[169] A device containing P3HT and compound **36** in a 1 : 1 ratio (by mass) was characterized by an energy conversion efficiency of 0.14%.[169]

The open-circuit voltage depends on the mutual position of the donor and acceptor energy levels[74] and can be varied by varying the HOMO and LUMO energies of a component of the photoactive layer.[170] The maximum attainable open-circuit voltage in organic photovoltaic devices is determined by the energy difference between the donor HOMO and the acceptor LUMO.[11,21] Having appropriately chosen substituents in the poly[4,8-bis-substituted-benzo[1,2-*b*:4,5-*b*′]-dithiophene-2,6-diyl-*alt*-3,4-disubstituted-thieno[3,4-*b*]-thiophene-2,6-diyl] (PBDTTT) derivatives **37a–c**, BHPS based on blends of these compounds with PCBM were prepared[171] and an open-circuit voltage of 0.76 V, a high short-circuit current of > 14 mA cm^{-2} and an energy conversion efficiency of 6.77% were attained.

An interesting method to improve the performance of polymeric BHPS is to incorporate selenium-containing heterocycles into the polymer chain.[172,173] For instance, incorporation of such heterocycles into the polyfluorene backbone causes a bathochromic shift of the absorption band relative to the corresponding absorption band of sulfur-containing analogues.[174] This enables a significant enhancement of absorption in the near-IR region without using metal complexes.[105]

Note that incorporation of unsubstituted fluorene into the conjugated chain also makes the colour much deeper owing to the developed π-system and high electronic symmetry of fluorene.[175] Additional deepening of the colour (bathochromic shift) is attained by introducing electron-

withdrawing groups at positions 2, 4, 5 and 7 of the fluorene molecule as a constituent of chromophoric systems.[176] Using nitro groups, absorption can be extended to the near-IR region (*e.g.*, 1000–1100 nm) for short-chain conjugated chromophores.[176–180] The phenomenon is due to efficient conjugation of nitro groups with the main chromophore, which leads to additional electronic transitions responsible for the new long-wavelength bands in the absorption spectra.

High-transparency polymeric media for photovoltaics were reported.[181] Earlier, numerous attempts were made to design BHPS based on conductive polymers, graphene and other materials with high transparency in the visible region.[182–186] However, the devices fabricated were of low transparency in the visible region and/or showed low energy conversion efficiency.[187–191]

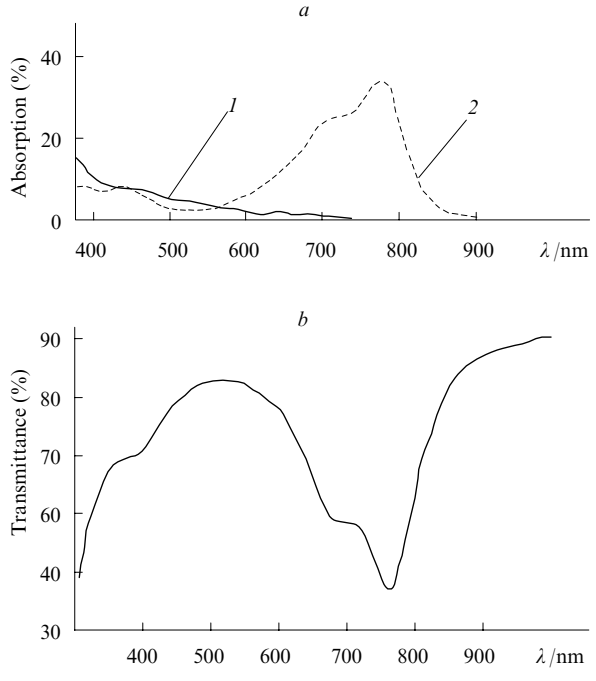
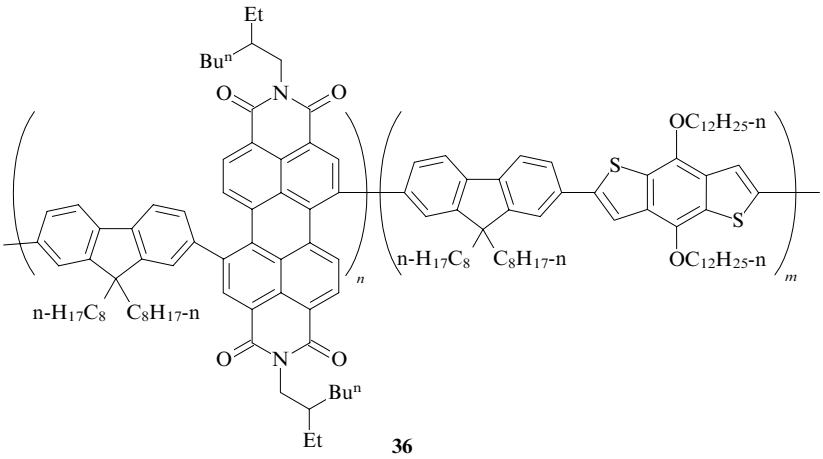
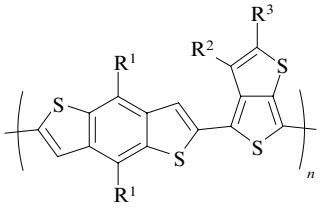


Figure 5. Absorption spectra (*a*) and transmission spectra (*b*) of the photoactive layer composed of polymer **38** and PCBM.[181] (*1*) Absorption of PCBM, (*2*) absorption of polymer **38**.

Structures 36, 37



36

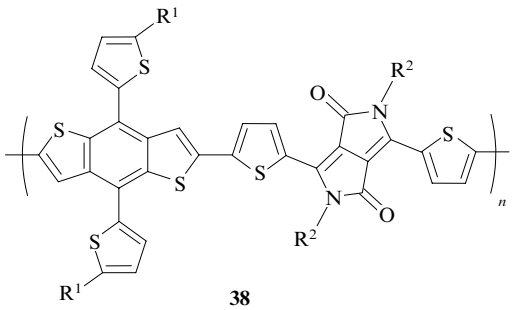


37a–c

$R^1 = OCH_2CH(Et)Bu^n$; R^2 = H:
$R^3 = CO_2C_8H_{17}$-n (**a**), C(O)CH(Et)Bu^n (**b**);
R^2 = F, $R^3 = C(O)C_7H_{15}$-n (**c**)

An electron-donor polymer **38** designed to fabricate transparent BHPS mainly absorbs in the near-IR region (Fig. 5 *a*).[181] A proper composition of the blend of polymer **38** with PCBM that mainly absorbs in the UV region provides a good transparency of 66% in the 550 nm region corresponding to maximum sensitivity of human eye (Fig. 5 *b*).[181] The BHPS includes two transparent electrodes and the active layer placed between them.

Structure 38



38

$R^1 = CH_2CH(Et)Bu^n$, $R^2 = CH_2CH(Bu^n)C_6H_{13}$-n

It should be noted that main problems in the design of transparent BHPS include the choice of the electrode material and the deposition procedure, especially in the case of the cathode (top electrode). The case in point is that the photoactive layer of a transparent BHPS is rather thin, soft and has poor adhesion to traditionally used metallic electrodes that are vacuum evaporated. To minimize the influence of these drawbacks, a composite cathode based on silver nanowire was proposed.[181] The cathode was spray coated onto the surface of the photoactive layer. Then, the silver nanowire was coated with TiO_2 to improve adhesion to the photoactive layer.[192] Also, a transparent conductive ITO filler was used as a component of the composite cathode in order to provide continuous contact with the photoactive layer. Here, it is necessary that the filler particles do not diffuse into the photoactive layer. The anode (bottom electrode) was fabricated from ITO. The photovoltaic parameters were determined for the cell illuminated from the top (type I BHPS) and from the bottom (type II BHPS). The results obtained are as follows:

Parameter	I_{sc}/mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
Type I BHPS	8.7	0.76	57.8	3.82
Type II BHPS	9.3	0.77	56.2	4.02

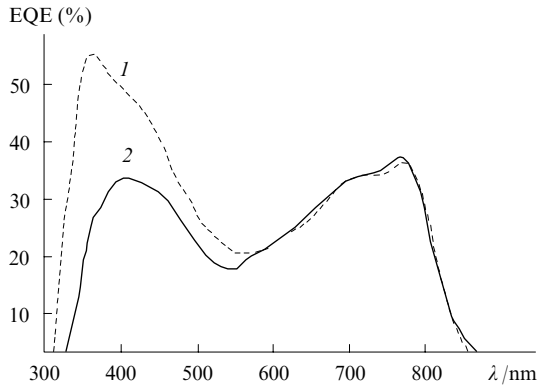


Figure 6. EQE spectra *vs*. wavelength of incident light for a BHPS composed of polymer **38** and PCBM: illuminated from the bottom (*1*) and from the top (*2*).[181]

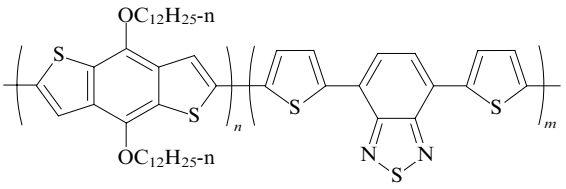
The EQE spectra of these BHPS are shown in Fig. 6.

The performance of transparent BHPS can be improved by placing a reflective mirror at the back of the transparent device to reflect the transmitted photons back to the device. Elaboration of this kind of transparent BHPS belongs to the key avenues of progress in photovoltaics; these devices can be components of smart windows and building-integrated photovoltaic energy converters.[181]

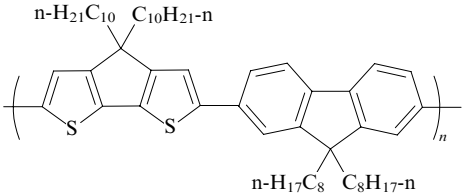
A number of donor-acceptor copolymers were synthesized and photovoltaic devices with the copolymer/PCBM active layer were fabricated.[193] A device based on the donor-acceptor copolymer **39** was characterized by the following parameters[193]:

I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
2.07	0.54	44.4	0.49

Structures 39, 40



39



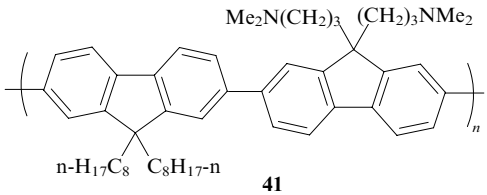
40

A novel conjugated copolymer **40** containing cyclopentadithiophene and fluorene moieties was used as donor in a BHPS with the **40**/PCBM active layer.[194] The energy conversion efficiency of the device was nearly 1.5%. The electronic and optical properties of copolymer **40** are very similar to those of the MDMO-PPV polymer **31b** that is widely used in photovoltaics (PCBM/**31b** blends provide an energy conversion efficiency of $>2.5\%$).[194] This suggests that optimization of the morphology of the **40**/PCBM photoactive layers may lead to similar efficiency values.[194]

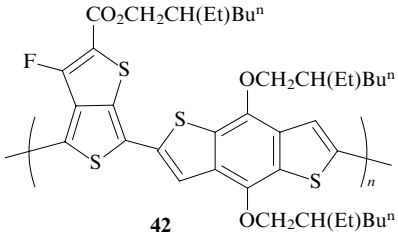
Inverted solar cells are of considerable interest. In these devices, charge carriers move in opposite directions as compared to their motion in conventionally used devices, *viz*., electrons move towards the bottom electrode (ITO) while holes move towards the top electrode.[195] In other words, ITO acts as a cathode and the top electrode becomes an anode.[55] The ITO cathode can be modified with metal oxides and carbonates, conjugated polyelectrolytes and other materials.[55] The top electrode (anode) is usually made of metals with high work function, such as gold, silver and copper.[195, 196] These electrodes can be deposited from colloidal solutions containing stabilized metal nanoparticles, thus avoiding energy-expensive and technically complicated vacuum deposition which is used for fabrication of conventional photovoltaic devices.[195] An undoubtful advantage of the inverted solar cells is that the top electrodes in them are made of metals with high stability to oxidation in air, whereas electrodes in conventional devices are less stable to oxidation which may lead to device degradation.[195, 196]

A certified energy conversion efficiency of $>9\%$ for inverted solar cells with a polymer/fullerene derivative active layer was reported.[55] The ITO surface was modified with conjugated polymer **41**.

Structures 41, 42



41



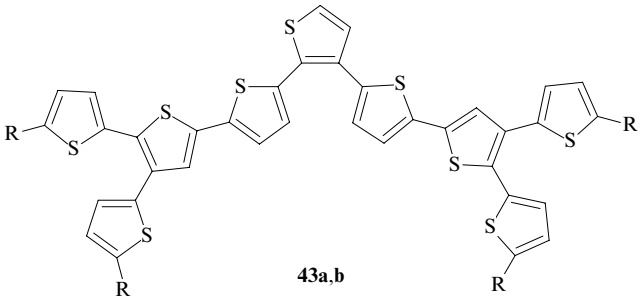
42

The active layer of the device was a blend of $PC_{70}BM$ with the low-band-gap semiconducting polymer **42**. The photovoltaic parameters of this device (type I BHPS) and a device with the same active layer and conventional architecture (type II BHPS) are as follows:[55]

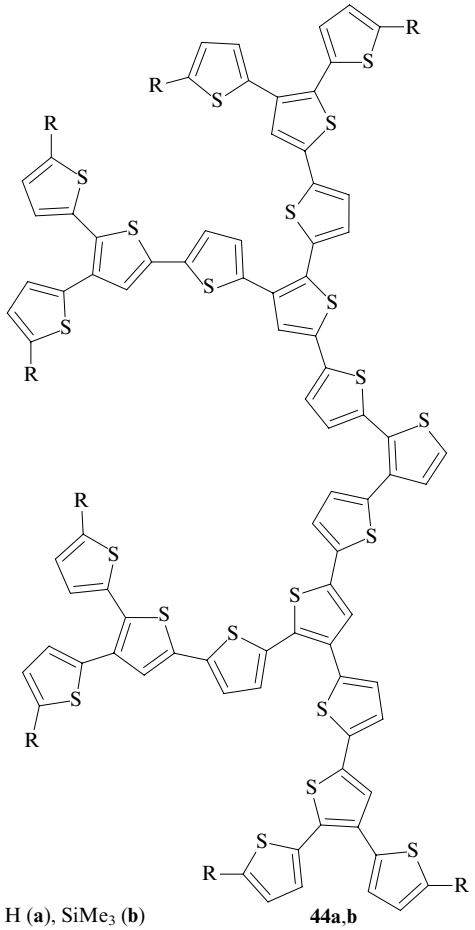
Parameter	I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)
BHPS I	17.46	0.75	69.99	9.2
BHPS II	15.4	0.76	70.6	8.24

Conjugated dendrimers form a new class of compounds that seem to be candidates for organic photovoltaics applications and have readily 'tunable' structures.[197] High solubility and good film-forming properties of conjugated dendrimers have made them basic components of promising materials for optoelectronic devices.[198, 199] Most often, conjugated dendrimers contain benzene,[200] carbazole[201] and thiophene[202] moieties. Thiophene-containing materials usually exhibit very good charge-transfer properties. A

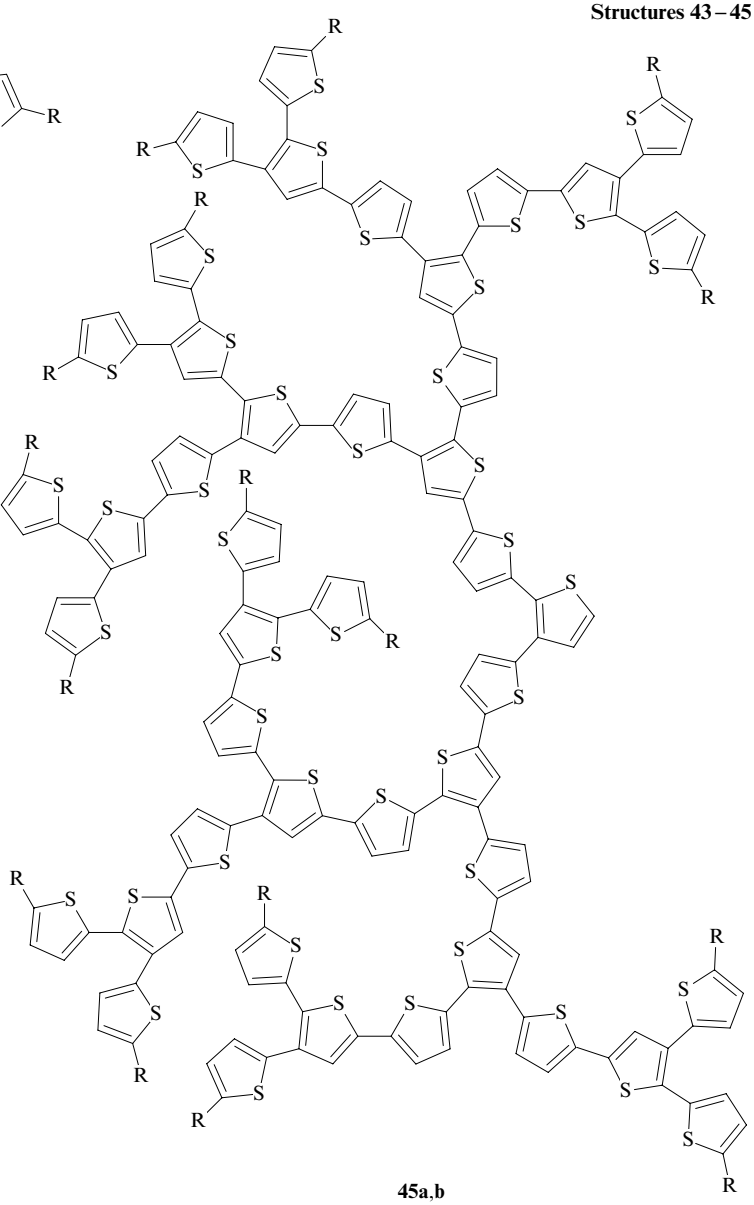
Structures 43 – 45



43a,**b**

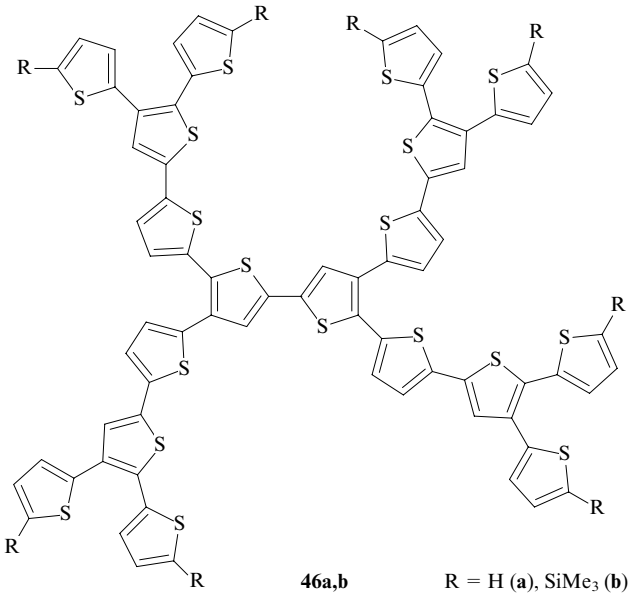


R = H (**a**), $SiMe_3$ (**b**) **44a**,**b**



45a,**b**

Structures 46



46a,**b** R = H (**a**), $SiMe_3$ (**b**)

number of dendritic and dendrimeric oligothiophenes **43a**,**b** – **46a**,**b** were used as electron donors in BHPS.[203] Dendrimer arms emerge from the common central fragment. A first-generation (G1) dendrimer has one branching point in each arm, a second-generation (G2) dendrimer has two branching points, *etc*. Two series of different-generation (from G2 to G4) dendrons and dendrimers were investigated.

The dendrimeric oligothiophenes in question have hyperbranched 3D structures and are readily dissolved in organic solvents (tetrahydrofuran, chloroform, chlorobenzene), thus facilitating the fabrication of photovoltaic devices. This class of thiophene-containing dendrimeric materials is of particular interest for BHPS applications because offers important advantages over traditionally used materials, namely:

— monodisperse composition and solubility with no solubilizing side chains lead to an increase in the absorption coefficient and to better transport properties;

— stable 3D structure specifies the lower bound of the domain size in the case of phase separation.[203]

Table 5. Parameters of BHPS based on dendritic and dendrimeric oligothiophenes and PCBM.[203]

Compound	I_{sc} /mA cm^{-2}	V_{oc}/V	FF (%)	η (%)	EQE_{max} (%)
43a	1.42	1.06	31	0.46	16
43b	1.53	1.03	29	0.46	18
44a	2.64	0.99	37	0.96	30
44b	2.10	0.93	34	0.66	24
45a	3.22	1.00	39	1.24	36
45b	3.29	0.99	34	1.12	38
46a	–	–	–	–	–
46b	2.39	0.94	35	0.79	28

The photovoltaic parameters of a series of BHPS containing various dendrimeric oligothiophenes are listed in Table 5.

VI. Effect of morphology of the photoactive layer on parameters of photovoltaic devices

The performance of photovoltaic devices depends strongly on the morphology of the active layer.[204, 205] Morphology optimization is an important task in the design of BHPS.[206–208] The optimized active layer should represent a nanocomposite with a domain size of the order of a few tens of nanometres, which is comparable with the diffusion mean free path of an exciton in an organic semiconductor.[209]

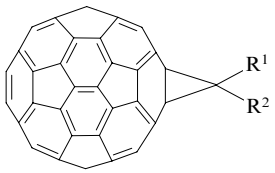
Morphology optimization in the course of solution deposition[210–213] allows one to eliminate subsequent thermal annealing of devices fabricated. For instance, the active-layer morphology of a structure based on P3HT/PCBM was optimized by varying the temperature of the casting solution.[213]

Transmission electron microscopy studies of the films thus obtained revealed the presence of P3HT nanofibres whose size and content depended on the temperature of the casting solution. At an optimal temperature of 45 °C, the content of the P3HT nanofibres was 42% and the photovoltaic parameters were I_{sc} = 9 mA cm^{-2} and η = 3.5%.[213]

Improvement of the photovoltaic characteristics of devices based on graphene derivatives also requires optimization of the morphology of their active layer by, *e.g.*, controlled thermal annealing. For instance, annealing of the structure P3HT/**29** at 160 °C led to improvement of the operating parameters. The active-layer morphology in this type of BHPS is significantly influenced by the amount of graphene **29**. An analysis of the film surface by scanning electron microscopy showed that the films containing 10% of graphene represent interpenetrating networks. This favours improved performance of the device. At a graphene content of <10%, individual graphene sheets were observed; at graphene concentrations >10%, large aggregates formed, but in both cases the device appeared to be less efficient.

Troshin *et al.*[51] synthesized 27 different derivatives of C_{60} fullerene (**6**, **47a–s**) and C_{70} fullerene (**48a–g**) which contained aliphatic and hetaryl substituents linked to fullerene by the cyclopropane moiety. These derivatives were used to fabricate BHPS. Electron donors were P3HT[51] and a number of other thiophene-containing conjugated polymers.[52] The physical properties and the measurable parameters of the structures were analyzed from the standpoint of solubility of the fullerene derivatives in the organic solvents used for film preparation. The influence of fullerene solubility on the film morphology was revealed.

Structures 47



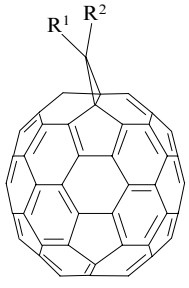
47a–s

Compounds **47**	R^1	R^2
a	$(CH_2)_3CO_2Me$	2-Th
b	$(CH_2)_3CO_2Me$	2-Fu
c	$(CH_2)_3CO_2Et$	Ph
d	$(CH_2)_2CO_2Me$	Ph
e	$(CH_2)_2CO_2Me$	4-$MeOC_6H_4$
f	$(CH_2)_2CO_2Et$	Ph
g	$(CH_2)_2CO_2Et$	2-Th
h	$(CH_2)_2CO_2Pr^n$	Ph
i	$(CH_2)_2CO_2Pr^n$	2-Th
j	$(CH_2)_2CO_2Pr^i$	Ph
k	$(CH_2)_2CO_2Bu^n$	Ph
l	$(CH_2)_2CO_2Bu^n$	2-Th
m	$(CH_2)_2CO_2Bn$	Ph
n	n-C_6H_{13}	Ph
o	$CO_2C_8H_{17}$-n	H
p	Bu^n	Ph
q	n-C_6H_{13}	2-Th
r	CO_2Et	$CO_2(CH_2)_2OMe$
s	$(CH_2)_2CO_2H$	$(CH_2)_2CO_2H$

The methanofullerenes used were structurally similar in that the cyclopropane moiety with two substituents was linked to the fullerene. Structural similarity also underlies the similarity between electronic spectra. The first reduction potentials of most fullerene derivatives under study were almost equal to that of PCBM.[214] This suggested that these methanofullerenes have close LUMO energies. Thin films of blends fullerene derivatives and conjugated polymers are most often cast from chlorobenzene. The composition and morphology of such donor–acceptor films depends to a great extent on the solubility of components.[215, 216] The performance of the BHPS fabricated using chlorobenzene is higher than that of the BHPS cast from toluene.[215]

The parameters of BHPS with the compositions P3HT/**6**, P3HT/**47** and P3HT/**48** obtained by cast from chlorobenzene are shown in Fig. 7. As can be seen, struc-

Structures 48



48a–g

Compounds **48**	R^1	R^2
a	Ph	$(CH_2)_3CO_2Me$
b	Ph	$(CH_2)_2CO_2Me$
c	Ph	$(CH_2)_2CO_2Et$
d	Ph	$(CH_2)_2CO_2Pr^n$
e	Ph	$(CH_2)_2CO_2Bu^n$
f	2-Th	$(CH_2)_2CO_2Pr^n$
g	2-Th	$(CH_2)_2CO_2Bu^n$

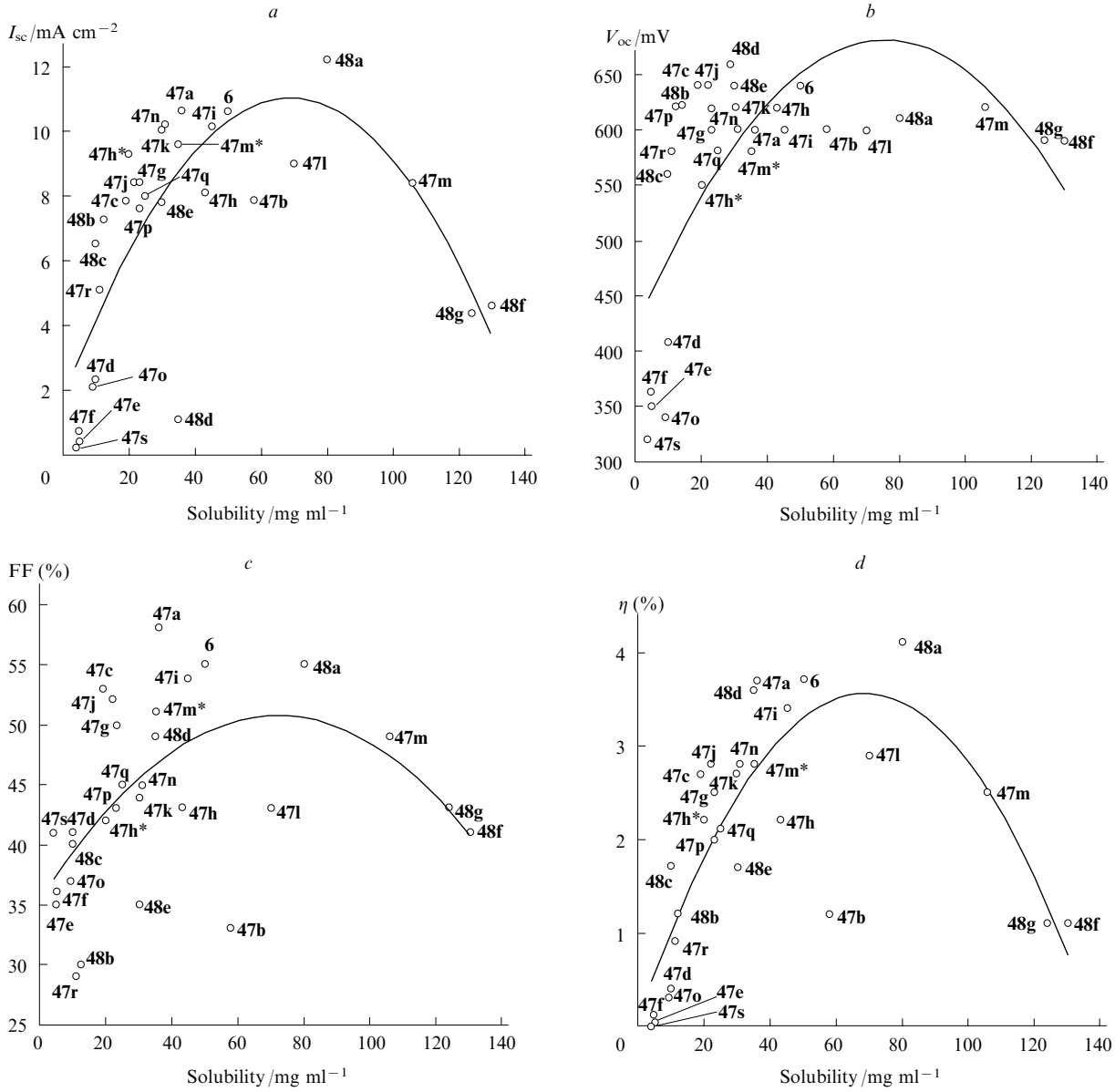


Figure 7. Plots of photovoltaic parameters (I_{sc}, V_{oc}, FF and η) *vs.* solubility of fullerene derivatives for BHPS with the compositions P3HT/**6**, P3HT/**47** and P3HT/**48**, measured after 15-min thermal annealing.[51]
Numbers given in bold are the numbers of compounds. Curves are guides to the eye.
The structures obtained by solution cast from toluene are asterisked; BHPS based on fullerene **47g** was prepared by solution cast from carbon disulfide.

tural similarity between fullerenes not necessarily leads to similar photovoltaic parameters; therefore, it is believed[51] that the parameters of the BHPS under study should be treated as being dependent on the solubility of the fullerene derivatives. The devices containing compounds **48a**,**b** are characterized by high short-circuit current because they contain C_{70} fullerene which absorbs light in a wider spectral range compared to C_{60} that is present in the related derivatives. Low performance of the BHPS based on compound **47s** was explained by the formation of unstable solvates with chlorobenzene, which decompose in the course of thermal annealing of the films. Rapid release of solvent vapours causes structural distortions in the film and foaming, thus leading to low performance of the device. An atomic force microscopy (AFM) examination[51] revealed domains of large aggregates on the surface of the films based on the fullerene derivatives characterized by poor solubility. Films fabricated from fullerenes with moderate solubility revealed moderate phase separation of P3HT and fullerene. The domain size was of the order of 5 to 15 nm, being comparable with the exciton diffusion length in these materials.[215] Blends of P3HT with the fullerenes well soluble in chlorobenzene (solubility >100 mg ml^{-1}) represent homogeneous solid solutions and show no phase separation. In these structures, charge transfer towards electrodes is difficult to occur and the efficiency of the device is rather low.

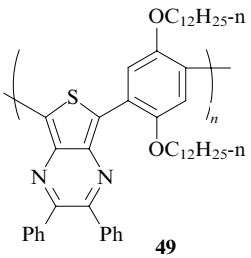
The morphology of the photoactive layer in the device based on compound **26** was optimized by introducing different amounts of 1-bromo-4-methoxybenzene to avoid

subsequent thermal annealing.[112] The best photovoltaic parameters were obtained for the device based on compound **26**, [6,6]-phenyl-C_{71}-butyric acid methyl ester ($PC_{70}BM$) and 1-bromo-4-methoxybenzene (2 vol.%).[111]

I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
7.81	0.96	50	3.6

The dependence of the efficiency of a BHPS based on poly(2,3-diphenylthieno[3,4-*b*]pyrazine-5,7-diyl-*alt*-2,5-didodecyloxy-1,4-phenylene) (PDDTP) (**49**) and PCBM on the degree of crystallinity of PDDTP was studied.[217]

Structure 49



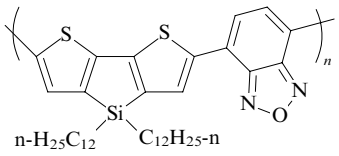
49

The photovoltaic devices thus fabricated were thermally annealed in a wide temperature range in inert atmosphere. A dependence was found of the short-circuit current I_{sc} on the annealing temperature, which exhibits a clearly defined maximum at 158 °C. Other photovoltaic parameters depend on the annealing temperature in less pronounced manner. It is believed[217] that the observed temperature dependence of the short-circuit current can be explained by partial crystallization of the polymer upon thermal annealing. To confirm this assumption, X-ray diffraction patterns of the samples mentioned above were obtained. They showed clearly seen crystallization peaks, the strongest peak being observed for the sample annealed at 158 °C. This was treated as an indication of clear correlation between the increase in the content of the crystalline phase in polymer **49** and the increase in the photovoltaic parameters. The short-circuit current was 5.51 mA cm^{-2}, the open-circuit voltage was 0.42 V and the energy conversion efficiency was 1.01%.

The morphology of the system **27**/$PC_{70}BM$ has strong effect on the performance of the device. An AFM study of these films revealed no phase separation on the surface despite a rather low solubility of compound **27** in chlorobenzene ($\sim$10 mg ml^{-1}; *cf.* $\sim$80 mg ml^{-1} for $PC_{70}BM$[113]). Probably, the large solubility difference causes a strong inhomogeneity of the films in the vertical direction, namely, after evaporation of the solvent during the formation of the photoactive layer the bottom layer will contain almost pure compound **27**, the content of $PC_{70}BM$ will increase from the bottom layer to the top and the top surface of the film will mainly contain $PC_{70}BM$. On the one hand, this architecture of the active layer is suitable for solving the problem of charge carrier recombination at the active layer–electrode interface. But on the other hand, the morphology of the active layer in bulk may appear to be unfavourable for efficient operation of the device. Probably, this is the reason for a rather low fill factor value for the device **27**/$PC_{70}BM$. To reveal the actual vertical morphology of the films, it is appropriate to study the cross-sections using specific microscopy techniques.[113]

A method for controlling the active-layer morphology of fullerene-containing BHPS with no thermal annealing[218] involves the addition of solvent to monitor phase separation in the course of film formation. The solvent should meet some criteria, *e.g.*, have a higher boiling point than the initial solvent; also, it is better to dissolve fullerene derivatives than the conjugated polymer donor[219–221] {in particular, poly[(4,4-didodecylthieno-[3,2-*b*:2′,3′-*d*]silol)-2,6-diyl-*alt*-2,1,3-benzoxadiazole-4,7-diyl] (**50**)}.

Structure 50



50

The HOMO and LUMO energies of polymer **50** determined by cyclic voltammetry are equal to −5.5 and −3.7 eV, respectively. The acceptor was $PC_{70}BM$. The polymer **50**:$PC_{70}BM$ ratio (by mass) was 1:2; otherwise, the performance of the device reduced. Experiments were carried out with two types of films. Type I films were cast onto the substrate from a chlorobenzene solution; type II films were cast from a mixture of chlorobenzene (98 vol.%) with 1-chloronaphthalene (2 vol.%). The photovoltaic parameters are as follows:

Parameter	I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
Type I films	4.3	0.7	54	1.6
Type II films	13.7	0.68	58	5.4

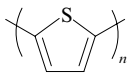
Significant differences between the photovoltaic parameters were explained[218] by different morphologies of the active layer of the devices. Type I films have the nonuniform surface covered with, *e.g.*, polymer aggregates, which is indicative of high phase separation and strong interaction between polymer macromolecules. This film structure is unsuitable for improvement of the operating efficiency of the photovoltaic device.[166] Films containing 1-chloronaphthalene had smooth surfaces, polymer **50** and $PC_{70}BM$ were uniformly distributed over the surface and no aggregates were observed. Thus, the results obtained in Ref. 218 provide a convincing proof that introduction of 1-chloronaphthalene precludes aggregation of the polymer, which leads to poorer phase separation and, as a consequence, improves the operating efficiency of BHPS.

Polymer/fullerene photovoltaic devices based on compound **12** with P3HT as a donor were reported.[77] Their energy conversion efficiency was only $\sim$2% *vs.* 4% for a P3HT/PCBM reference cell despite the fact that the devices have similar optical and electrochemical properties. This may be due to different morphologies of the active layer. AFM studies revealed an increase in the grain size on the surface of the **12**/P3HT film after thermal annealing because raising the temperature leads to a considerable increase in the mobility of molecules **12** and subsequent formation of large domains of fullerene **12** with a lower surface energy.[77] Also, coarse-grained morphology of the **12**/P3HT film is due to lower solubility of compound **12** compared to PCBM. With this active-layer morphology, the efficiency of hole transport can decrease because of the loss of

excitons generated within large domains of fullerene **12**.[222,223] Therefore, improvement of the properties of the photovoltaic devices based on compound **12** requires novel approaches to optimization of the active-layer morphology.[77]

The efficiency of photovoltaic devices was improved by introducing a polythiophene buffer layer between PEDOT : PSS and the P3HT/PCBM active layer.[224] Polythiophene (**51**) is insoluble in most of organic solvents and the polythiophene buffer layer precluded charge carrier recombination at the anode/active layer interface.[225,226] The component ratio in the active layer was 1 : 1 by mass. A thin polythiophene buffer layer was deposited electrochemically. The thickness of the electrodeposited film of polymer **51** appeared to be dependent on the charge density. The photovoltaic parameters of devices with buffer layers of different thickness proportional to the charge density were determined.[224]

Structure 51



51

A comparison of the results obtained for the devices with buffer layer and those obtained for the reference cells containing no buffer layer revealed significant improvement of photovoltaic parameters upon introducing the buffer layer. The measured parameters of samples with the polythiophene buffer layers of different thickness are listed in Table 6.

AFM studies showed that at a charge density of 0.2 mC cm^{-2}, the layer of polymer **51** has an island structure at the top of the PEDOT : PSS layer. As the charge density reached a value of 0.4 mC cm^{-2}, the polythiophene film completely covered the surface of the PEDOT : PSS layer and was rather smooth. As the current density further increased to 1 mC cm^{-2}, the polythiophene layer became rather thick, inhomogeneous and its surface became rough; this led to poorer photovoltaic parameters of the device. Charge carrier recombination was best precluded by a homogeneous smooth buffer layer in the form of polythiophene film; this made it possible to achieve an energy conversion efficiency of 4.11%. An important role in the formation of the active-layer morphology was also played by the PEDOT : PSS polymer belonging to 'synthetic metals'.[227] It levels the surface, is a good hole injector, exhibits readily variable conductivity and is characterized by high transparency.[228–233] Owing to the planar structure of PEDOT, polymer **30** is characterized by high electron delocalization along the backbone. It was stated[232] that PEDOT : PSS films are composed of 'conductive' or 'polaron' PEDOT units incorporated into a quasi-insulating PSS matrix and that charge transport occurs jumpwise or through tunneling between the polaron islands. Polymer **30** has good contact resistance for holes that move from one PEDOT island to another through the insulating PSS matrix. Spatial variation of conductivity may occur in the films.

Table 6. Parameters of BHPS based on P3HT/PCBM containing a polythiophene buffer layer electrochemically deposited at different charge densities (Q).[224]

Q /mC cm^{-2}	I_{sc} /mA cm^{-2}	V_{oc} /V	FF (%)	η (%)
Reference sample	8.77	0.6	66.5	3.5
0.2	11.61	0.59	58.6	4.01
0.4	12.32	0.59	56.5	4.11
0.6	12.14	0.58	55.1	3.88
0.8	12.51	0.60	44.0	3.31
1.0	11.74	0.58	40.8	2.78

Electrical parameters of PEDOT : PSS may vary over a wide range. Hole-injection layers in organic photovoltaic devices are often created using PEDOT : PSS layers with moderate conductivity ($\sim 10^{-3}$–10^{-5} S cm^{-1}). An original product[228] had a conductivity of $\sim$ 10 S cm^{-1} which could be increased tenfold by adding polyhydroxyl compounds, *e.g.*, sorbitol, glycerol, or using a polar solvent like *N*-methylpyrrolidone. The increase in conductivity is due to subtle changes in the structure of dry PEDOT : PSS particles, especially, in their interaction. Strengthening of intra-chain interactions is detected from changes in the optical anisotropy upon reorientation of polymer chains upon the addition of sorbitol. Highly conductive ($>$1400 S cm^{-1}) PEDOT : PSS was reported.[234] The high conductivity allows polymer **30** to be utilized as an anode instead of ITO in photovoltaic devices. It was stated[228] that the actual conductivity depends strongly on the microscopic structure of dry particles in the film, which depends on the annealing conditions and on the distribution of PEDOT islands in the matrix specified by the polymerization conditions. An advantage of PEDOT : PSS consists in the possibility to obtain smooth films from aqueous dispersions by centrifugation.

Air-stable PEDOT : PSS films are characterized by a work function of 5.2 eV. PEDOT : PSS is suitable to extend the operating lifetime of photovoltaic devices because it hampers oxygen diffusion from ITO into the active layer.[229] This polymer is widely used as a material for antistatic coatings, electric contacts and electrodes for organic semiconductor devices.[228–232]

VII. Conclusion

According to Academician Zh I Alferov, the winner of the 2000 Nobel Prize in physics, the photovoltaic method of solar energy conversion to electricity is the most efficient method despite the fact that now it accounts for only 2% of the overall amount of energy produced worldwide. It is assumed that photovoltaics will help in solving the energy challenges of the future.[25]

Modern trends in the field of BHPS mainly include improvement of their performance and search for simpler fabrication technologies. Here, of paramount importance is targeted design of novel materials for organic photovoltaics, *viz.*, fullerene derivatives,[51,57,235] conjugated polymers,[55] dendrimers[203] and organic dyes with strong absorption in the near-IR region.[48,84] Among promising materials for organic photovoltaic structures, there are graphene derivatives that are suitable for diverse applications including anodes,[150] cathodes,[151,152] components of the active layer[130] and act as hole injectors.[134] Combining versatility and transparency of polymers will make it possible to fabricate fully organic devices containing no inorganic glasses, ITO, metallic electrodes, *etc*. In addition to search for novel materials, there are some other topical problems:

— fabrication of active layers with the desired morphology [206, 207] to improve the efficiency of transport of photo-induced charge carriers;

— facilitation of methods for optimization of the active-layer morphology [218] in order to simplify the fabrication of photovoltaic devices.

Thus, attainment of the maximum possible efficiency of BHPS requires a complex approach that combines application of novel materials and mandatory control of the active-layer morphology.

To extend the operating life of materials for BHPS, considerable attention should be paid to methods for minimization of photo- and electrochemical degradation [68] that accompanies the operation of photovoltaic devices.[236]

In his lecture at the St Petersburg Scientific Forum, Alan Heeger, the winner of the 2000 Nobel Prize in chemistry, said that an efficiency of 13% was achieved for modern solar cells based on polymeric materials [25] (*cf.* more than 24% for silicon cells [1]). This is one of a few drawbacks of organic photovoltaic structures.

Progress in modern solar power engineering requires the design of novel and optimization of the available photovoltaic materials. A promising approach seems to be the use of printing technologies for production of flexible BHPS.[237–240] This makes it possible to significantly reduce the production expenditures, but the technologies should be improved. Recently, intensive studies have been carried out on hybrid organo-inorganic photovoltaic structures based on 3-alkyl-substituted polythiophenes and TiO_2,[241–243] CdSe,[244] $CuInSe_2$.[245] They exhibit rather high performance.[246–248] Relatively simple and cheap methods for improvement of their photovoltaic parameters include the introduction of buffer layer [249, 250] fabricated from inorganic salts and oxides [251–255] and the addition of carbon nanotubes to polymer/fullerene solar cells for better hole transport.[256–258]

Tandem polymer solar cells make it possible to achieve an energy conversion efficiency of 10.6%.[259]

Studies on materials based on conjugated polymers and nanoparticles,[260] as well as carbon nanotubes with titania [261–267] or zinc oxide,[268–273] unsubstituted polythiophene,[274] copolymers based on cyclopentadithiophene [194, 275] and benzodithiophene [276] are of great importance.

The design of conjugated polymers containing heavy (bromine, iodine) atoms is of considerable interest. The presence of heavy atoms leads to enhanced intersystem crossing and generation of triplet electron–hole pairs that form charge carriers much more readily than corresponding singlet pairs and, therefore, enhance the photovoltaic effect.[277]

The search for novel sensitizing dyes for photovoltaics remains topical,[278–284] especially in connection with the development and practical application of new types of electrodes. Novel *N*,*N*′-diphenylperylenediimides and other perylene derivatives are also promising because of high photochemical stability and intense absorption in the red and near-IR regions.[285–289] Studies on the photovoltaic properties of novel donor-acceptor systems based on borodipyrromethene dyes are now in progress.[290, 291]

Yet another class of candidate materials for BHPS includes polymeric composites containing polymethine dyes, either ionic [292] or intra-ionic (squarylium [284, 293] of merocyanine [294–297]) ones. This is due to the fact that polymethines exhibit the widest range of photophysical and photochemical properties among organic dyes.[87, 88, 298] Also, ionic character of polymethines allows them to produce a local electric field within the photovoltaic structure; this improves the dissociation of excitons and, as a consequence, causes the photocurrent to increase. However, one should take into account the possibility of photocurrent hysteresis to occur, which was reported recently.[295] A large number of possible structural modifications allows the properties to be 'adjusted' to photovoltaics applications. In addition, merocyanines and squaraines are interesting in connection with the BHPS applications because they can be introduced into the photoactive layer by both cast and evaporation techniques.[294–299] Triphenylamine dyes,[300, 301] organometallic compounds,[302] carbon nanotubes [303–306] and copper phthalocyanine nanorods [307, 308] find ever-growing use.

Researchers are interested not only in novel synthetic, but also natural dyes. Photoprocesses occurring in the latter should be studied in detail in order to produce the photovoltaic effect. In this connection, successful application of natural dyes, namely, anthocyanes [309] and novel porphyrin derivatives [114, 310, 311] in photovoltaics should be pointed out.

Studies on transparent photovoltaic media are also promising.[181] They will be aimed to search for polymers and dyes characterized by the maximum possible absorption in the UV and near-IR regions and minimal absorption in the visible region.

Targeted search for novel dyes and polymers requires extension of experiments on the determination of the HOMO/LUMO energies by cyclic voltammetry methods.[114] This is necessary for correct parameterization of modern quantum chemical methods for calculations of complex organic molecules and their absorption spectra (because the results of energy calculations are often strongly different from experimental values) and for subsequent theoretical predictions.[312–314]

J-Aggregates of organic dyes form a unique class of solar energy converters. They are characterized by maximum intensity of absorption, weak electron-phonon coupling, the ability to transfer the energy of exciton excitation along the *J*-aggregate chain[315–324] and may appear to be candidates for applications in BHPS. In addition, high surface concentration of *J*-aggregates will help to minimize the size of photovoltaic devices.

A great potential of the new branch of science and technology called plasmonics still remains unused in organic photovoltaics applications.[325, 326] The case in point is that metal (especially, gold and silver) nanoparticles exhibit excellent optical properties and can amplify electromagnetic radiation through plasmon resonance,[327–333] which is characterized by very high Q-factor and can be tuned with ease in a wide spectral range by varying the size and shape of nanoparticles, thus being very promising for enhancement of the photovoltaic effect.[328] This is favoured by the recently developed methods for coating inorganic nanoparticles with shells from organic dyes[330, 334] including their *J*-aggregates.[335] The latter can contribute largely to the resonance properties of plasmons and quantum dots.[335, 336]

Quantum dots are first of all promising in that their photophysical and electrophysical properties can be varied in targeted manner by varying the particle size and

shape.[337, 338] Also, quantum dots can provide absorption of solar energy in a much wider spectral range compared to conjugated organic compounds. High photochemical stability of quantum dots will ensure long operating lifetime of photovoltaic devices.[337, 338]

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