

Plastic Solar Cells: from Basic Research to Devices

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Addition of fullerenes to π -conjugated polymers results in an electron transfer process from the conjugated polymer onto the fullerenes upon illumination. The photoinduced electron transfer in these composites is ultrafast with a quantum efficiency approaching unity. The separated charges are metastable. This photoinduced electron transfer may be utilized in interesting applications like photodiodes and solar cells.

The discovery of semiconducting, conjugated polymers and the ability to dope these polymers over the full range from insulator to metal resulted in the creation of a class of new materials that combines the electronic and optical properties of semiconductors and metals with the attractive mechanical properties and processing advantages of polymers. Many conjugated polymers (Figure 1a) in their undoped, semiconducting state are electron donors upon photoexcitation (electrons promoted to the antibonding π^* band).

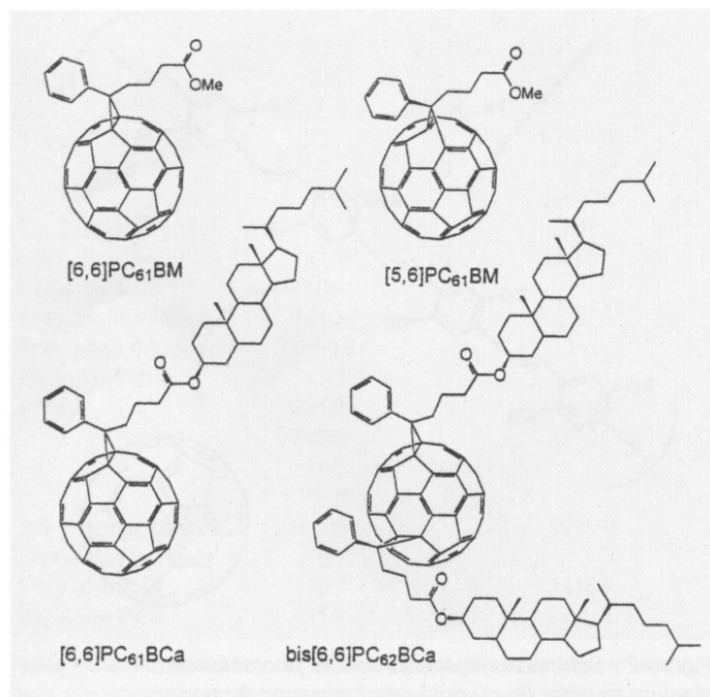


Figure 1b - Molecular structure and abbreviation of fullerenes

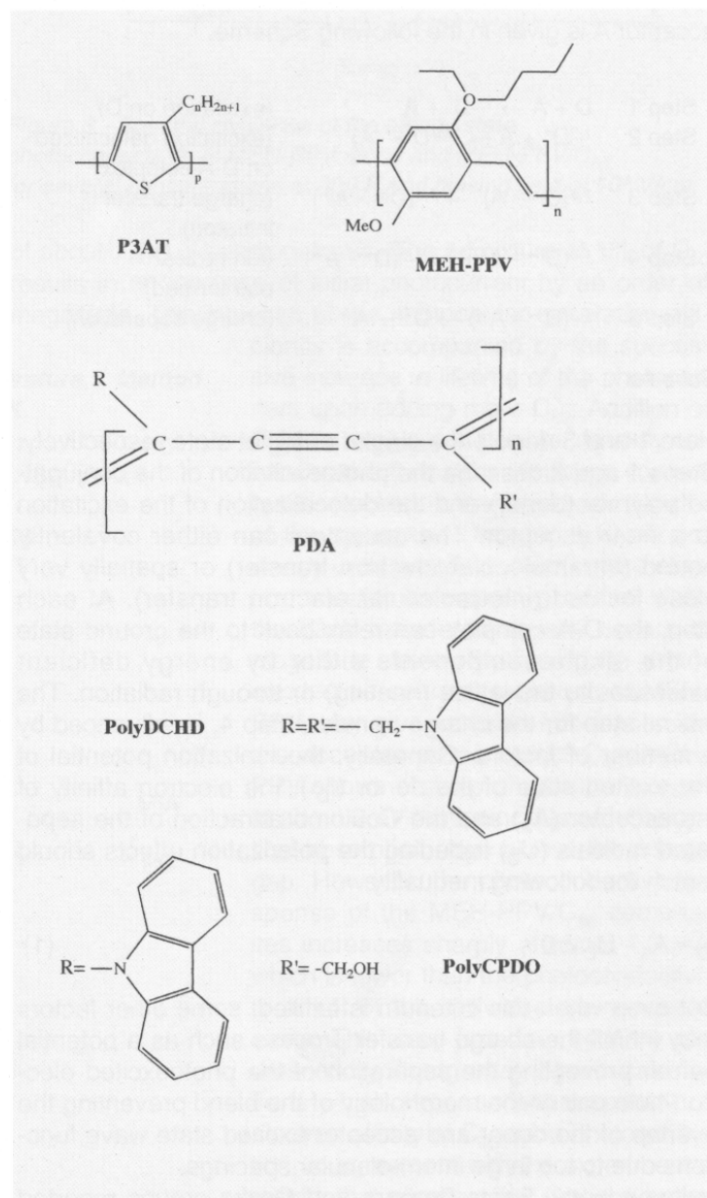
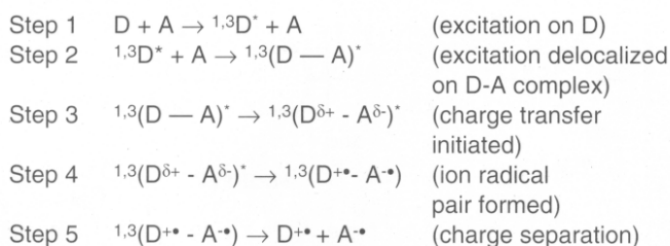


Figure 1a - Molecular structure and abbreviation of common conjugated polymers

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The possibility to utilize strongly light absorbing systems to duplicate the overall process of photosynthesis is attracting more and more the scientific community. Conjugated polymers seem to fulfill all properties required for photosynthesis: strong light absorption and charge separation. The idea of using this property in conjunction with a molecular electron acceptor to achieve long living charge separation was based on the stability of the photoexcitations (such as polarons or polaron pairs) on the conjugated polymer backbone. After the transfer of the photoexcited electron to an acceptor unit, the resulting cation radical (positive polaron) species on the conjugated polymer is stabilized due to the delocalization of the excitation over several repeating units. In analogy to the chemical doping process, we will describe the photoinduced electron transfer as "photodoping". A basic description of the intermolecular electron transfer between a donor D and an acceptor A is given in the following Scheme.



Scheme

Here 1 and 3 denote the singlet or triplet state respectively. Steps 1 and 2 describe the photoexcitation of the conjugated polymer (donor) and the delocalization of the excitation to a near acceptor. The acceptors can either covalently bound (intramolecular electron transfer) or spatially very close located (intermolecular electron transfer). At each step, the D-A complex can relax back to the ground state of the single components either by energy deficient processes to the lattice (heating) or through radiation. The critical step for the charge transfer, step 4, is influenced by a number of factors. Generally, the ionization potential of the excited state of the donor (I_{D^*}), the electron affinity of the acceptor (A_A) and the Coulomb attraction of the separated radicals (U_C) including the polarization effects should match the following inequality:

$$I_{D^*} - A_A - U_C < 0 \quad (1)$$

But even when this criterium is fulfilled, some other factors may inhibit the charge transfer process such as a potential barrier preventing the separation of the photoexcited electron-hole pair or the morphology of the blend preventing the overlap of the donor and acceptor excited state wave functions due to too large intermolecular spacings.

Independently, Santa Barbara and Osaka groups reported studies on the photophysics of mixtures and bilayers of conjugated polymers with fullerenes [1, 2]. The experiments clearly evidenced an ultrafast, reversible, metastable photoinduced electron transfer from conjugated polymers onto the C_{60} in solid films. Schematic description of this phenomenon is displayed in Figure 2. Using this molecular effect at the interface between bilayers consisting of semiconducting polymer (poly(2-methoxy,5-(2'-ethyl-hexoxy)-*p*-phenylene) vinylene,

hereafter referred to as MEH-PPV) and C_{60} films, diodes were demonstrated with rectification ratios on the order of 10^4 which exhibited a photovoltaic effect. Soon after the detection of the ultrafast electron transfer between conjugated polymers and C_{60} , functionalized, highly soluble fullerenes were available (Figure 1b). The superior solubility of these fullerenes compared to C_{60} enabled the fabrication of highly fullerene concentrated composites. Furthermore, significant improvement of the relatively low collection efficiency of the D/A bilayer has been achieved by using phase separated composite materials through control of the morphology of the phase separation into an interpenetrating network. Monochromatic power conversion efficiency of solar cells made from MEH-PPV/fullerene composites was subsequently increased by two orders of magnitude to approximately 3% [3]. Recently, the Cambridge group reported monochromatic power conversion up to 6% for laminated solar cells made by phenyl substituted and cyano substituted PPV [4].

In this article, we will review experimental results to discuss the photophysical properties of conjugated polymer/fullerene composites. Strong emphasis will be given to the photovoltaic application potential of these D/A composites. In reference to the photoinduced electron transfer phenomena we discuss the role of a special class of polydiacetylenes recently synthesized with a photoconductive side group: the polycarbazolyldiacetylenes.

The photophysics of conjugated polymer/fullerene composites

PPV derivatives

We review here the photophysical effects on properties of conjugated polymers upon blending with fullerenes. The optical absorption spectra of a MEH-PPV/ C_{60} film with different C_{60} content compared to the optical absorption spectrum of the components alone show a simple superposition of the two components. Furthermore, the positions of the absorption

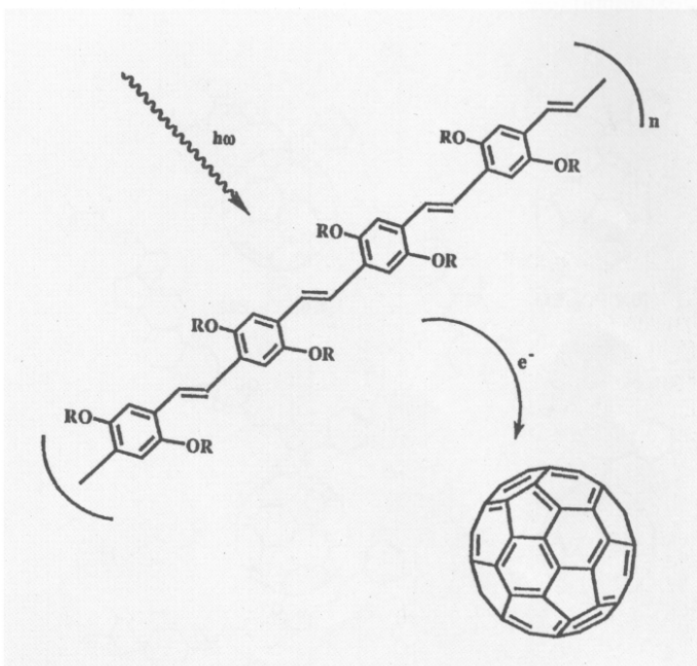


Figure 2 - Schematic illustration of the photoinduced electron transfer from conjugated, semiconducting polymers onto Buckminsterfullerene, C_{60}

maxima of the single components in the composite are not shifted compared to the pristine materials. There is no indication of states below the π - π^* gap of the conducting polymer as might arise from interactions like ground state doping between the two materials. Upon addition of small amounts of fullerenes the strong photoluminescence of MEH-PPV is quenched by a factor in excess of 10^3 . The luminescence decay time is reduced from $\tau_0=550$ picoseconds to $\tau_{rad} < 60$ picoseconds (the instrumental resolution) indicating the existence of a rapid quenching process; e.g. subpicosecond electron transfer [1]. Similar results were obtained for poly(3-alkylthiophene) (P3AT) by the Osaka group [2]. The fast electron transfer process is also confirmed by the quenching of the intersystem crossing to the triplet manifold and by the presence of strong infrared active vibration (IRAV) bands in the photoinduced absorption measurements. In addition, ESR measurements under illumination of the sample, show the presence of two photoinduced radicals which have been identified as a positive polaron on the polymer chain and C_{60}^- .

One of the most remarkable effects of the photoinduced charge transfer from the conjugated polymers to fullerenes is the sensitization of the photoconductivity. Figure 3 shows, on a semilog plot, the spectral response of the cw-photoconductivity (cw-PC) of MEH-PPV alone and MEH-PPV/ C_{60} composite for different concentrations of C_{60} . These room temperature data are normalized to the constant incident photon flux

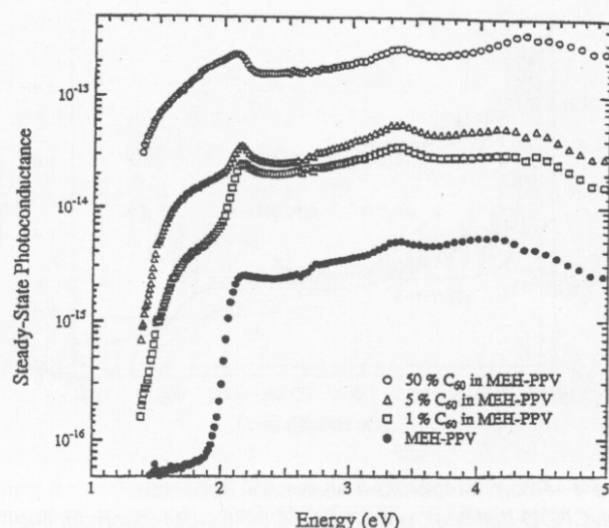


Figure 3 - Spectral response of the steady state photoconductivity of MEH-PPV alone and MEH-PPV/ C_{60} for several concentrations at 300 K and biasing field of 10^4 V/cm

of about 7.5×10^{14} photons/cm²s. The admixture of 1% of C_{60} results in an increase of initial photocurrent by an order of magnitude. This increase of the photocarrier generation efficiency is accompanied by the successive increase in lifetime of the photocarriers upon adding more C_{60} . Addition of C_{60} to a conjugated polymer not only enhances the formation of charge carriers but also prevents the rapid back transfer of the separated charges. It is still not clearly understood why the forward electron transfer is happening so fast while the back transfer is several orders of magnitude slower. Studies on the photoconductive spectral response of MEH-PPV may give valuable information on the acceptor properties of C_{60} . MEH-PPV shows a sharp photocurrent onset at about 2 eV that coincides with the optical absorption edge across the energy gap. However, the photoconductive response of the MEH-PPV/ C_{60} composites increases sharply at about 1.3 eV, which is lower than the photoconductivity onset of the individual components alone. This issue has been recently addressed by Zhang *et al.* [5] with calculations which show an enhancement of electron affinity of C_{60} upon photoexcitation, in agreement with the experimental results that photoexcited C_{60} is a better electron acceptor compared to C_{60} in the ground state. These findings may explain the remarkably enhanced photoconductivity over the broad spectral range from the near infrared to the ultraviolet. Although the effect of the C_{60} on the mobility of the carrier should be considered (the mobility will probably de-

Material	Mobility, μ (cm ² /Vs)	$\mu_{ } / \mu_{\perp}$	Electric Field, V/cm	Temperature, K	Method
PDA-TS	5		3×10^4	RT	PC
	8		$\sim 10^4$		
	0.1-10	10^2	$\sim 10^3$		
	10^{-4}		2×10^7	148	
	3		3×10^4	RT	
	1.2-5		8×10^3		
	5	$\sim 10^3$	3.5×10^4		
	5		3×10^4		
	10	$\sim 10^3$			
	3.5 ± 1.5	$\sim 10^3$	4×10^3		
8.9		10^4			
0.8-3		$10^4 - 10^5$			
7.6				MIA	
Trans (CH) _x	0.11-3.2	2-140			PC
	1		1.5×10^4	RT	
	2	50	1.5×10^4		
0.75-5.3		2×10^4			
C_{70} (single crystal)	1		10^5		
C_{60}	2×10^{-2}		3×10^4		
Polysilane	10^{-4}				
Poly-methyl-silane	7×10^{-3}				
Poly-alkyl-thiophene	$10^{-7} - 10^{-4}$				
Exa-thiophene	5×10^{-9}				
PPV	2×10^{-4}		2.5×10^3		
	2×10^{-2}		2×10^5		
	10^{-4}				
	$10^{-5} - 10^{-6}$				
2,5-Dieptyloxy-PPV	6×10^{-5}		5×10^3		
Diphenyl-PPV	10^{-4}				
Phenyl-PPV	10^{-4}		3×10^4		
Dialkoxy-PPV	5×10^{-7}				

Table - Carrier mobilities for different conjugated polymers and fullerenes. Data are obtained both from photoconductivity (PC) measurements or from photoinduced microwave absorption (MIA)

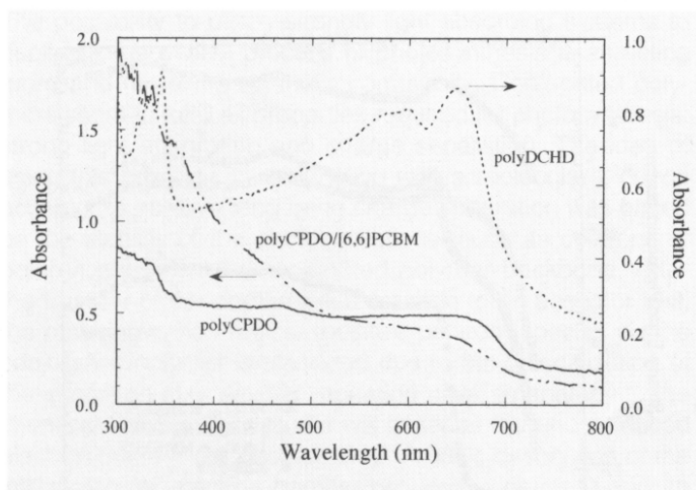


Figure 4 - Room temperature absorption spectrum of polyCPDO (full line), polyCPDO/[6,6]PC₆₁BM (dash dot line) and polyDCHD (dashed line)

crease as a result of the deep trap sites associated with C₆₀ clusters), the significant effect of C₆₀ on the charge carrier generation efficiency is evident from the observation that even 1% C₆₀ in the polymer matrix enhances the cw-PC by more than an order of magnitude. This observation is in full agreement with the photoinduced electron transfer phenomenon which leaves metastable positive polarons on the polymer backbone after the electron transfer; *i.e.* "photodoping".

Polydiacetylenes

Another important result from the photophysical studies on conjugated polymer/fullerene composites is the complete absence of photoinduced interaction between fullerenes and red form polydiacetylenes [6]. Spectroscopic results from absorption-emission spectroscopy, transient and cw photoinduced absorption studies as well as photoconductivity studies show, that in contrast to the high quantum efficiency photoinduced electron transfer definitely established in the PPV as well as P3AT composites with C₆₀, photoinduced electron transfer from red phase PDA to fullerenes is absent. For the PDA, the structure and morphology of the composite films might be relevant to the absence of the photoinduced electron transfer. In studies on P3AT and solubilized PPV films, heavily loaded with C₆₀ so that partial phase segregation occurs, photoinduced electron transfer has been observed. On the other hand, the complete absence of the electron transfer in red phase PDA-4BMCU/[6,6]PC₆₁BCa composites cannot be explained by sample morphology alone. Comparison of the ionization potentials of the PDAs (around 5.5 eV) are very close to that of P3ATs and PPVs (5.1-5.2 eV). Thus the striking difference between these two systems of conjugated polymers must have its origin in the photophysics of the red form polydiacetylenes. As pointed out before, the Coulomb attraction between the electron and the hole must be overcome (step 4). For the PDAs investigated the electron-hole (exciton) binding energy is relatively large (0.4-0.5 eV). Indirectly, these results imply, that the Coulomb interaction in P3ATs and PPVs is sufficiently screened in comparison with the PDAs.

Even though these results seem to indicate that PDAs are not a class of conjugated polymers suitable for the study of the electron transfer process to fullerenes, recent studies on the photoexcited states of polycarbazolyldiacetylenes, and in

general, of blue form PDAs [7, 8], indicate that the situation could be more favourable when the blue form of PDAs is used as donor. PDAs give a very interesting chance to the organic chemists to synthesize materials with controlled electronic states. Due to the higher crystallinity of PDAs with respect to the other conjugated polymers, higher values for the carrier mobility are expected (see Table). The insertion of different side groups allows the modification of the interaction between the conjugated polymer and the environment, thus providing a control of the processability of the material. Moreover, the side groups affect the electronic states of the backbone, in particular their excited states that are the ones most involved in the photophysics of the materials. In our laboratories we studied a particular class of PDAs where the photoconductive carbazolyl side groups are directly or indirectly (through a methylene spacer), symmetrically or asymmetrically attached to the backbone. This molecular engineering strategy allows the preparation of materials with very interesting electronic properties. In Figure 4 we report the absorption spectrum of polyDCHD, polyCPDO, and polyCPDO/[6,6]PC₆₁BM mixture. The absorption spectrum of polyDCHD shows the typical features of the ordered blue form of PDAs with an excitonic peak at about 650 nm followed by its vibronic progression. Below 350 nm the characteristic absorption bands of the carbazolyl group appear. The absorption spectrum of polyCPDO is quite different from that of polyDCHD. In fact this spectrum exhibits no negligible absorbance in the full

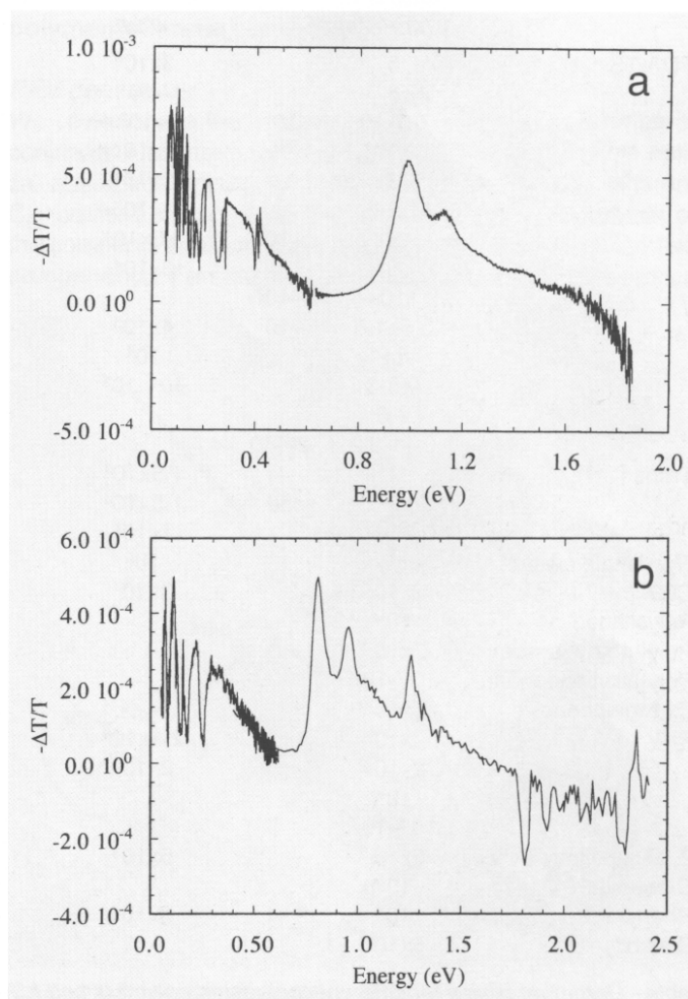


Figure 5 - Photoinduced absorption spectrum of polyCPDO (a) and polyDCHD (b)

range 200-700 nm. In other words, the absorption spectrum of polyCPDO unlike that of polyDCHD gives a better match to the solar spectrum. Moreover the action spectrum of the photoconductivity in polyCPDO clearly shows that charges are photogenerated when photons, whatever is their energy, are absorbed [10] in contrast to the typical case of PDAs where the presence of an excitonic state below the conduction band prevents the photogeneration of charges for photon energies below the interband gap which is situated around 2.5 eV (3 eV) for the blue (red) form. The absorption spectrum of polyCPDO/[6,6]PC₆₁BM seems to be the superposition of the absorption spectrum of the two isolated molecules.

Another interesting feature of these materials is their photoinduced absorption (PIA) spectra. In Figure 5 we report the full spectral range PIA spectra for polyCPDO (a) and polyDCHD (b). The PIA spectrum of polyDCHD shows bands at 0.82, 0.96 and 1.26 eV. In addition a broad band at about 0.1 is observed with superimposed "windows" corresponding to IRAV modes. These bands are originated by charged "defects" which break the local symmetry and therefore make Raman modes infrared active [11]. It was shown that the bands at 0.1 and 0.82 eV are due to photogenerated charged states (polarons or bipolarons) and that the band at 1.26 eV is due to weakly trapped triplet excitons. The band at 0.96 eV is a vibronic replica of the 0.82 eV signal [8]. The presence of the IRAV modes unambiguously shows that the photexcitation process of the material generates charged excited states. It was shown that the photogeneration of charged states in PDAs is typical of their blue form [8] that is of the form achieved when

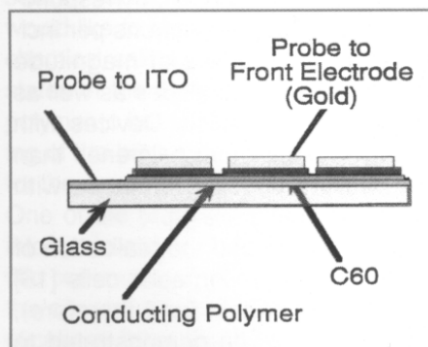


Figure 7 - Schematic cross section of the heterojunction devices fabricated from MEH-PPV and C₆₀

the conjugated chain is fully extended and highly ordered. From the point of view of the photovoltaic applications, the PIA spectrum of polyCPDO seems more interesting because it shows only photoexcited charged states.

Indeed, in this polymer, no signal associated with triplet excitons could be detected. This characteristic seems to be typical of polycarbazolydiacetylenes with the carbazolyl group directly bound to the conjugated skeleton. Only asymmetrically substituted PDAs could be obtained since symmetrical substitution of aromatic rings directly attached to the skeleton prevents the polymerizability of the monomers. These results stimulated us to further study the photogeneration process of the charged excited states in PDAs and in particular of the charge transfer process on which the photovoltaic devices are based. To this end we studied the PIA spectra of polyCPDO mixed with [6,6]PC₆₁BM. In Figure 6 the PIA spectrum of polyCPDO/[6,6]PC₆₁BM is

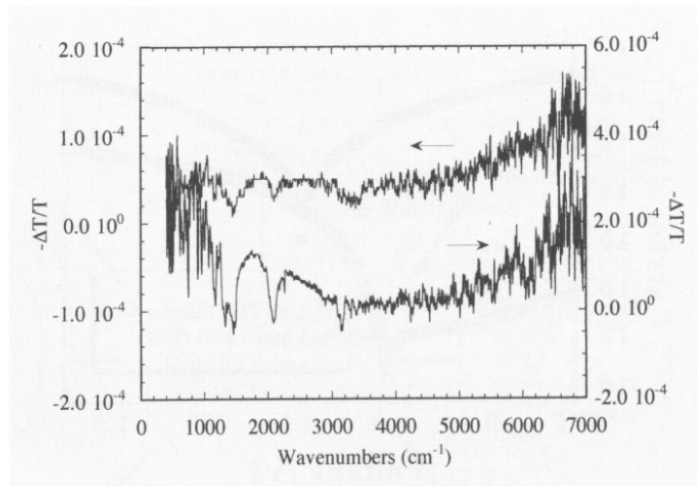


Figure 6 - Comparison of the medium infrared photoinduced absorption spectra of polyCPDO/[6,6]PC₆₁BM (left scale) and polyCPDO (right scale)

reported and compared with that of pure polyCPDO. The spectrum looks very similar to that previously reported for polyCPDO alone, even though it is more noisy due to the disorder caused by the partial phase segregation between the two components. We would like to stress that the data here reported for PDAs must be considered only preliminary because good quality samples of polyCPDO/[6,6]PC₆₁BM as required for the study of the charge transfer processes could not be obtained so far. For this reason no quantitative evaluation of the influence played by [6,6]PC₆₁BM can be performed. Further work is necessary to clarify this point.

In addition to PIA measurements, LESR studies, i.e. the differential ESR spectra obtained by subtracting the ESR spectra with light on and off, which are able to distinguish between spinless (bipolarons) and spin 1/2 (polarons) photoexcitations are in progress. While we detect photoinduced spin 1/2 radicals from polyCPDO, photoinduced C₆₀⁻ is not detected in polyCPDO/C₆₀ composites [12]. We are currently working to improve the morphology of polyCPDO/[6,6]PC₆₁BM. A very promising approach to increase the miscibility of polyCPDO with C₆₀ could be given by a new polyCPDO with long alkyl chains in the 3,6 positions of the carbazolyl rings, recently synthesized.

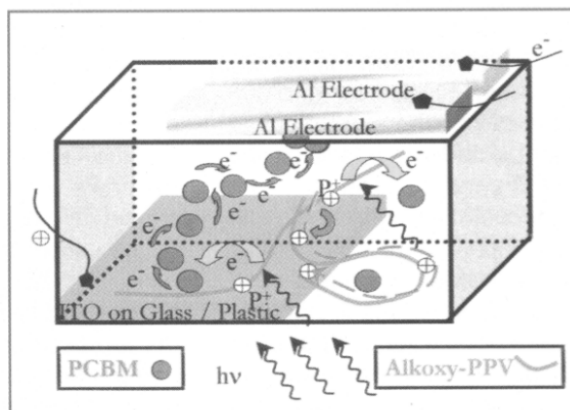


Figure 8 - Schematic illustration of the interpenetrating bicontinuous donor/acceptor (conjugated polymer/fullerene) network of internal heterojunctions. Green full dots are fullerenes molecules, brown ribbons are conjugated polymer chains

Photovoltaic devices from conjugated polymers - fullerenes composites

The utilization of organic materials for photovoltaic devices has been investigated intensely during the last couple of decades. Due to the fact, that research was limited to small molecules as conjugated polymers were not available at this time, bilayers were the preferred device geometry. Tang demonstrated photovolta-

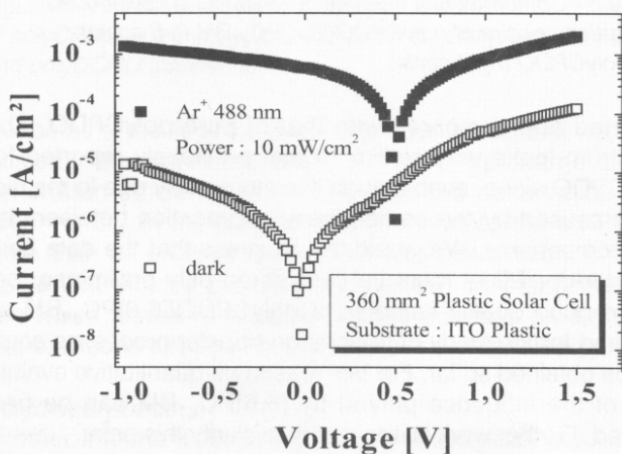
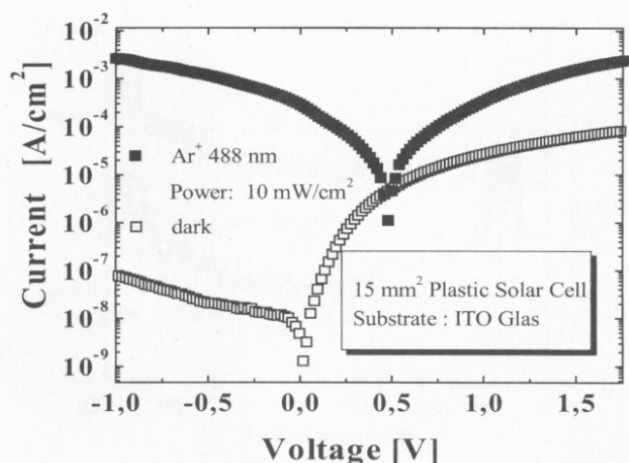


Figure 9 - Current voltage characteristics of a Al/MEH-PPV:[6,6]PC₆₁BM/ITO-glass device (15 mm²) compared to a Al/MEH-PPV:[6,6]PC₆₁BM/ITO-flexible transparency device (360 mm²)

ic activity in small molecular bilayers that were vacuum evaporated [13]. Extensive literature exists on the fabrication of solar cells based on small molecule dyes as well as donor-acceptor systems. Inorganic oxide semiconductors have also been used to facilitate electron transfer from organic dyes to achieve charge separation and photovoltaic conversion. With the improvement and development in the synthesis of soluble conjugated polymers, conjugated polymer layers have been used in solar cells. Even the acceptor properties of fullerenes have been utilized for photodiodes, although not with conjugated polymers. Yamashita and coworkers reported a bilayer photodiode based on the organic donor tetrathiafulvalene (TTF) and C₆₀ [14]. Because of the ultrafast photoinduced electron transfer with long lived charge separation, the conjugated polymer/C₆₀ system offers a special opportunity. Using conjugated polymers as donors with different acceptors, results in photoinduced charge separation with quantum efficiency near unity and with correspondingly enhanced device performance. Semiconducting devices with asymmetric contacts, that is a low work function metal on one side and a high work function metal on the opposite side, function as a "tunneling injection diodes"; such devices have been described by Parker [15]. A schematic cross-sectional view of such devices is displayed in Figure 7. Devices made with pure conjugated

polymers and asymmetric contacts have their main application in electroluminescence diodes (LED). Such a device can also be utilized as photovoltaic cell, however energy conversion efficiencies were typically 10⁻³-10⁻²%, too low to be used in practical applications [16].

Investigation of the conjugated polymer/fullerene bilayers showed the great potential of the ultrafast photoinduced electron transfer for photovoltaic devices. However, the small charge separation area at the interface limits the efficiency of this device geometry. On the other hand, all photophysical studies of conjugated polymer/fullerene composites showed, that even 1% C₆₀ influences the photophysics in mixed films. Consequently, interpenetrating phase separated D/A network composites would appear to be ideal photovoltaic materials. Through control of the morphology of the phase separation into an interpenetrating network, one can achieve a high interfacial area within a bulk material. Since any point in the composite is within a few nanometers of a D/A interface, such a composite is a "bulk D/A heterojunction" material. If the network is bicontinuous, as shown schematically in Figure 8, the collection efficiency can be equally efficient. The realization of devices with these bulk D/A heterojunctions showed important progress for photovoltaic applications [3]. The short circuit current, I_{sc}=0.5 mA/cm² under 20 mW/cm² illumination, corresponding to a collection efficiency of η_c= 7.4% electrons per incident photon [3] approximately two orders of magnitude higher than that of pure MEH-PPV tunnel diodes as well as of the MEH-PPV/C₆₀ heterojunction device. Devices with high fullerene concentration (4 times more fullerenes than conjugated polymer) showed the best performance with power conversion efficiencies around 2.9% [3].

The characterization of these devices and the realization of large scale, flexible network heterojunction solar cells [16] are promising. Recently the first ultrathin (and therefore), flexible all plastic solar cells have been demonstrated to yield comparable efficiencies as small area cells on glass. Figure 9 shows a comparative study on the efficiency of small area glass and large area flexible cells. Upscaling does not seem to influence the performance. Further optimization of device performance can be achieved by optimization of the device physics:

- i) optimize the metallic electrodes to achieve good ohmic contacts on both sides for collection of the oppositely charged photocarriers;
- ii) optimization of the bandgap to the solar spectrum (1.35 eV);
- iii) optimize the charge carrier mobility of the single components as well as transport properties of the network.

For the field of organic photovoltaics, the synthesis of new materials allows to optimize their electronic properties. As shown before, polyCPDO absorption spectrum match the solar emission spectrum better than other conducting polymers. This behavior, which is peculiar of polyCPDO with respect to other PDAs is caused by the direct attachment of the aromatic group to the conjugated skeleton. An increase of the electronic delocalization is achieved thus pulling the π-π* transition into the NIR. Moreover, conjugated polymers with low band gaps, such as poly(isothianaphthene) and derivatives may bring further reduction of the bandgap. As discussed before, the order achieved in PDAs samples, probably due to their solid state polymerization, gives rise to films with long travelling path for the charge carriers. This proper-

ty, together with the strong photogeneration of charged carriers, results in increased interest to study the photophysics of polyCPDO in order to learn, whether polyCPDO may be a candidate for photovoltaic applications.

Because of the advantages that would be realized with polymer-based photovoltaic devices, such as low cost fabrication in large sizes and in desired shapes with mechanical flexibility, efficient "plastic" solar cells would have a major impact.

Conclusions

The discovery of an ultrafast, reversible, metastable photoinduced electron transfer in conjugated polymer/fullerene composites has opened a large area of scientific and technological interest. In this report we limited the topic on the utilization of the photoinduced electron transfer for photodiodes and photovoltaic devices. However, the application potential of conjugated polymer/fullerene composites is much larger. Some examples for future research topics are shown in Figure 10. We would like to stress the importance of quantum chemical calculations of the electronic excited states of the composite for the design of proper conjugated polymer to be synthesized in the near future. The understanding of the basic physical and chemical properties is important for the development of devices based on new materials. One of the promising properties that justify the fast growing interest in conjugated polymer/fullerene devices by the scientific community as well as by industry is the processability of the conjugated polymers. Photodetector and photovoltaic applications have reached the stage where development work on an industrial level is beginning. Upscaling of small area device production up to endless role to role manufacturing seems to be realized in near future, as the basic processing knowledge is already present from work with conventional polymers. Thus, the photophysics and the associated device applications of the conjugated polymer/fullerene composites imply an important scientific and industrial opportunity.

Acknowledgements

This paper is dedicated to the memory of Umberto Bianchi, Professor of Macromolecular Chemistry, University of Genova. The work on PDAs here presented is a part of a fruitful collaboration with Prof. C. Dell'Erba, C. Cuniberti and G. F. Musso. Support from MURST is acknowledged. ♦

Glossary

*	excited state
A:	acceptor
AA:	electron affinity of the acceptor
cw:	continuous wave
cw-PC:	cw-photoconductivity
D:	donor
I:	ionization potential
IRAV:	infrared active vibration
MEH-PPV:	poly(2-methoxy,5-(2'-ethyl-hexoxy)-p-phenylene)vinylene
P3AT:	poly(3-alkyl-thiophene)
PDA:	polydiacetylene
PDA-4BCMU:	poly[5,7-dodecadiyn-1,12-diol-bis(4-butoxycarbonylmethylurethane)]
PIA:	photoinduced absorption
polyCPDO:	poly[1-(N-carbazolyl) penta-1,3-diy-5-ol]
polyDCHD:	poly[1,6-di(N-carbazolyl)-2,4-hexadiyne]
PPV:	poly(phenylene)vinylene
UC:	Coulomb attraction of the separated radicals

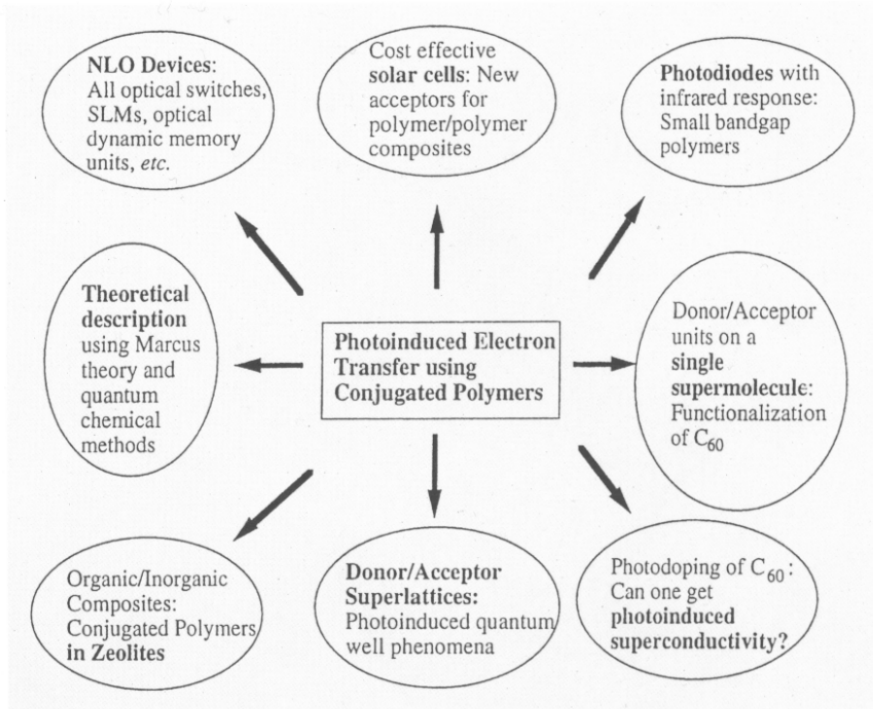


Figure 10 - Some examples of current and future scientific research directions utilizing the ultrafast, reversible, photoinduced electron transfer from conjugated polymers onto fullerenes

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