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Perspective

Semiconducting Polymeric Materials

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(Semi)conducting polymers with a π-conjugated (hetero)aromatic backbone are capable of transporting charge and interact efficiently with light enabling their utilization in a variety of opto-electronic devices. In this report and in the additional papers of this special issue, several classes of π-conjugated polymers are reported and reviewed. Furthermore, an introduction to field-effect transistors (FETs), polymer light-emitting diodes (PLEDs), and organic photovoltaic cells (OPVs) as well as a summary of the state-of-the-art organic semiconductors used in the different devices are provided.

Keywords semiconducting polymers, organic field-effect transistors, organic photovoltaic cells, organic light-emitting diodes, conjugated polymers

1. Introduction

Polymers or plastics have been traditionally considered passive materials and the electronic industry has been extensively using them as insulators of metallic conductors, photoresists, and incapsulation layers.[1] However, since the serendipitous discovery in 1977 that chemical doping of polyacetylene (PA, Fig. 1) resulted in a highly conducting material by as much as eleven orders of magnitude compared to the pristine polymer,[2,3] many academic and industrial research laboratories have initiated projects in this exciting area. The era of "electro-active" or "conducting polymers" began. Although the initial research and development emphasis was to enable highly conducting materials as a replacement of conventional metals, more recent efforts targeted their semiconducting properties. Furthermore, the focus has shifted from synthesizing insoluble and untreatable powders such as polyacetylene and unsubstituted poly(heterocycles) such as polythiophene to enhancing the performance of semiconducting polymers exhibiting far

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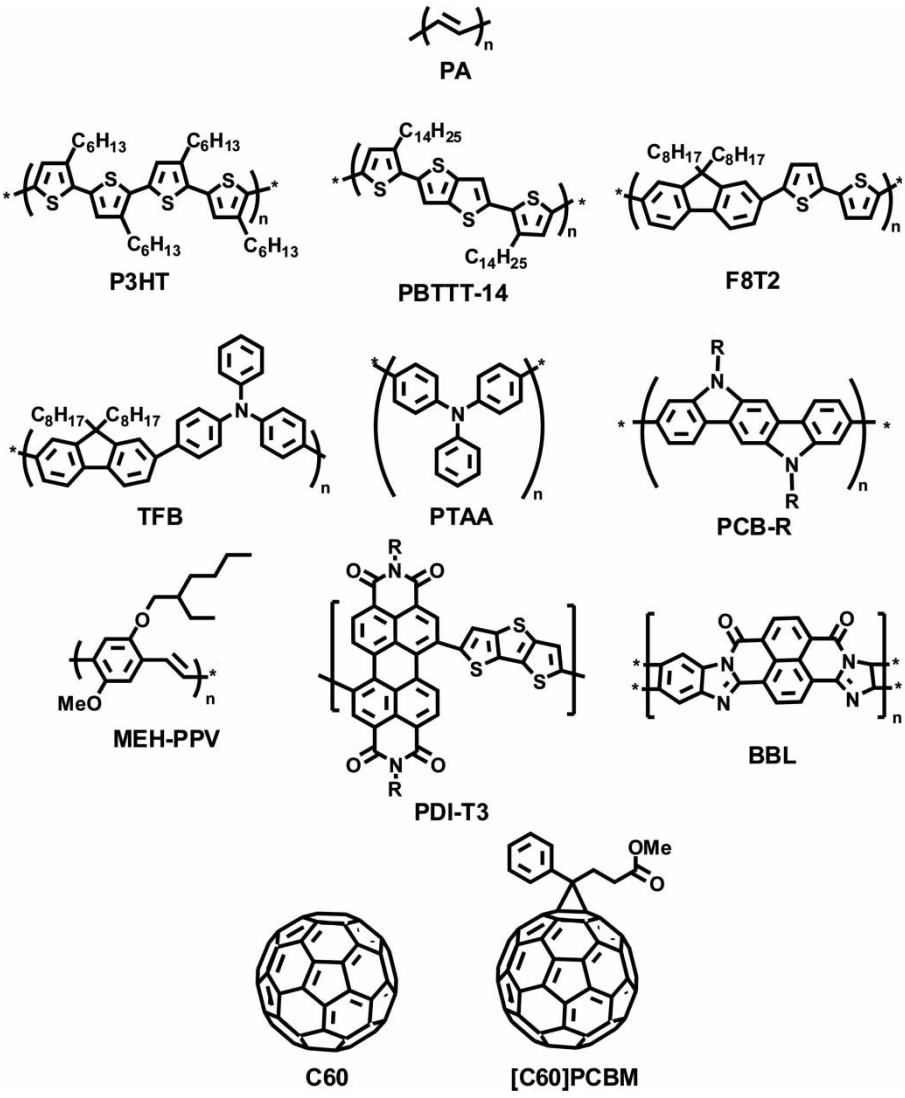

Figure 1. Chemical structure of various semiconducting polymers and fullerenes.

greater solubility and manufacturability such as MEH-PPV and P3HT (Fig. 1). The net result is that during the last 25 years an extensive library of polymers has been created by designing and synthesizing new polymerizable monomeric structures exhibiting physical and chemical properties tuned for implementation in a variety of opto-electronic devices. These modifications ultimately affect the charge transport characteristics of the bulk solid and define the role that the material may play in various device configurations. Examples of the applications include, but are not limited to, conducting elements (after proper doping), organic photoconductors, field-effect transistors (FET),[4–8] light-emitting diodes (LED),[9–12] photovoltaic cells (PV),[13–15] sensors,[16,17] lasers (solution[18] and solid state[19–22]) photodetectors,[23] and organic circuits[24] for integration into display technologies and low-cost electronics.[25] The main advantages of using the solution-processable

polymers lie in inexpensive materials processing and high throughput device assembly mainly via printing methodologies. Properly designed polymers may be processed via solution techniques, thereby eliminating the need for expensive lithography and vacuum deposition steps necessary in silicon-based microelectronics. Low-temperature solution processing also enables the utilization of inexpensive substrates such as flexible plastics in combination with processing options such as spin casting, photolithography,[26,27] ink jet printing,[28–30] soft lithography,[31] screen printing,[32] and micromolding[33] onto almost any type of substrate, including flexible ones.[34]

In the following paragraphs we will very briefly describe three very exciting device applications where (semi)conducting polymers are currently investigated. Figure 2 shows their device structure and operation principles.

2. Field-Effect Transistors

Field-effect transistors (FETs) were among the first semiconductor-based devices fabricated with a conducting polymer. A FET is a simple device composed of three contacts (source, drain, and gate), a dielectric layer, and a semiconducting layer (Fig. 1a). FETs act essentially as electronic valves, by modulating the semiconductor channel conductance via the gate field. In this device structure the current flow between the source and the drain electrodes is controlled by the application of two independent biases, one between the source and the drain (V_d) and one between the source and the gate (V_g). The latter bias controls the carrier density in the FET channel, hence it determines whether the device is switched "off" ($V_g = 0$ V, low carrier density) or switched "on" ($V_g \neq 0$ V, high

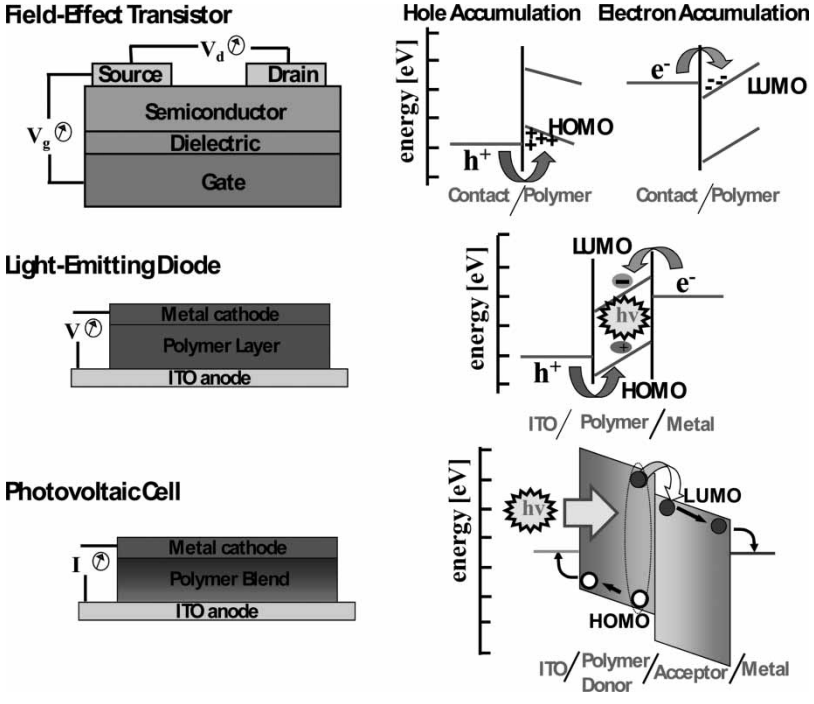

Figure 2. Structure and energy diagram of various semiconductor-based devices namely the field-effect transistor, the light-emitting diode and the photovoltaic cell.

carrier density). Among the organic semiconductor classes used in OFETs, polythiophenes are among the most extensively investigated due to their chemical/electrochemical stability and their ready functionalization.[35] The first report of a polyheterocycle-based FET utilized a polythiophene,[36] and poly(3-hexyl)thiophene (P3HT)[37] being the first truly high-mobility polymeric material. Since then, utilization of various heterocycles, chemical modification(s) of the conjugated core, variations in ring-to-ring connectivity and substitution pattern, have resulted in the synthesis and evaluation of a large number of polymers. Note that the majority of polymeric semiconductors for FETs can efficiently transport hole (p-channel semiconductors) whereas the electron-transporting polymers (n-channel semiconductors) for FETs are far more rare.[38]

Among the p-channel polymers, P3HT is doubtless the most investigated and P3HT–based FET performance strongly depends on various parameters including polymer purity, regioregularity, doping,[39] and molecular weight[40] as well as P3HT film deposition solvent,[41] film morphology,[42] film thickness,[43] and the fabrication process.[44] For instance, when regioregular P3HT (>90% of head-to-tail linkages) is used to fabricate FETs, a dramatic increase in mobility is observed relative to regiorandom poly(3-alkylthiophene)s. Highly regioregular P3HT also forms lamellae with an edge-on orientation (π-π stacking direction in the plane of the substrate) when spun from chloroform. Mobilities of 0.05 to 0.1 cm^2/Vs were obtained for 96% regioregular P3HT.[45] In contrast, spin-coated films of P3HT with low regioregularity (81% HT linkages) consist of lamellae having a face-on orientation (π-π stacking direction perpendicular to the substrate) and exhibit low mobilities of $\sim 10^{-4}$ cm^2/Vs. Another important thiophene-based polymer for FETs is PBTTT, which exhibits charge-carrier mobilities as high as 0.2–0.6 cm^2/Vs for devices annealed under nitrogen, and up to 0.7 cm^2/Vs for devices with a 5 μm channel length.[46]Another interesting polymer employed in FETs is poly[9,9-dioctylfluorene-co-bithiophene] (F8T2). F8T2-based FETs have been fabricated using inkjet printed solution-processed polymer electrodes (water-soluble poly[3,4-ethylenedioxythiophene] doped with polystyrene sulfonic acid (PEDOT/PSS)), insulators (polyimide), and an active organic semiconducting layer of F8T2.[30] It was demonstrated that F8T2, which is a nematic liquid crystalline conjugated polymer semiconductor, can be preferentially oriented by rubbing polyimide layers, and when used as the active channel in FETs, exhibits a mobility of 0.02 cm^2/Vs and an on/off ratio of 10^5.[47] Other interesting p-channel polymeric semiconductors are those based on 9,9-dialkylfluorene-alt-triarylamine (e.g., TFB),[48] carbazole (e.g., PCB-R),[49,50] and triarylamine (PTAA) units.[51]

As mentioned above, n-channels polymers are rare.[38] A co-polymer of *N,N′*-dialkyl-1,7-dibromo-3,4,9,10-perylene diimide with dithienothiophene (PDI-T3) is one of the most promising n-channel polymer to date.[52] This polymer was found to be soluble in chloroform, THF, and chlorobenzene and could readily be processed from solution. The molecular weight of x was not very high and ~15 kD using GPC (polystyrene standards). PDI-T3-based FETs (with Al source/drain electrodes) exhibit electron mobilities as high as ~ 0.01 cm^2/Vs. Unfortunately, this material does not function in ambient atmosphere. The only known air-stable n-channel polymer for FETs is BBL, which exhibit a carrier mobility of ~ 0.1 cm^2/Vs.[53] The drawback of this polymer is the limited processability in conventional organic solvents, which may be overcome by proper core substitution.

3. Light-Emitting Diodes

In 1990 the Cambridge group led by Richard Friend reported the emission of light from a semiconducting polymer sandwiched between two contacts and connected to a battery.[54]

The discovery of electroluminescence (EL) in conjugated polymers, i.e., the emission of light upon electron-hole recombination as the result of an electric current flown, has provided a new impetus for the development of polymer light-emitting diodes (PLEDs). These elements are fundamental for display and other applications.[55] In PLEDs and in OLEDs (organic LEDs), the latter being their small molecule-based equivalents, the holes and electrons injected from the device contacts recombine and produce luminescence with a wavelength (color) dictated by the energy difference between the molecular excited and ground states. For the majority of conjugated polymers, an electron injection is more difficult than a hole injection, since conventional polymers are more easily oxidized than reduced, resulting in an unbalanced charge injection into the device. This problem was solved by using metals with a low work function (e.g. calcium) as the cathode as well as other interfacial engineering using electron-injection layers. However, since calcium is highly susceptible to atmospheric degradation, this contact can be covered with a metal that is not sensitive towards oxygen and moisture, like aluminum. With the appropriate choice of an emissive polymer, contact materials, and a device design, PLEDs exhibiting high external quantum efficiencies have been obtained, with efficiencies that are comparable with the best EL devices based on inorganic materials. Turn-on voltages of 5 V or below have also been achieved by the use of charge-transporting layers, enabling devices to be run from low-power sources like batteries. Polymer-based electroluminescent displays provide a good alternative to the well-established display technologies based on cathode-ray tubes and liquid-crystal displays (LCDs) with respect to processability and viewing-angle. Especially for the application in large-area displays and flexible displays, for which the conventional methods are not well suited, polymer light-emitting diodes may offer great advantages.

4. Photovoltaic Cells

Two years after the PLED breakthrough at Cambridge, the Santa Barbara group reported the first results on polymer-based photovoltaic cells,[56,57] of which the principles can be regarded as the inverse of the EL process. In photovoltaic (PV) devices, bound electron–hole pairs (excitons) are created upon illumination. To convert the absorbed light into electricity, the excitons must first dissociate into the separate charges which then have to be collected at the device contacts (holes at the anode and electrons at the cathode). This process is possible thanks to the use of contacts with different work functions enabling the formation of a built-in potential field within the device. To describe exciton dissociation, the concept of an electron donor and an acceptor is frequently used, in which the electron affinity of the electron acceptor material in the PV blend should be larger than the ionization potential of the donor component.[58] Since the initial investigations, most PV cells were based on blends of poly(*p*-phenylene vinylene) (PPV)[59] until the thermal treatment of the PV cell led to very high efficiencies for poly (3-hexylthiophene) (P3HT) as the light absorber/electron donor/hole conductor material. The polymeric materials are combined with a substituted C_{60} derivative (PCBM) as the electron acceptor/electron conductor.[60–62] PPV-based and P3HT-based materials are widely investigated for their opto-electronic properties[63–65] and the ability of PCBM to accept several electrons[66] makes it particularly attractive for use as an electron acceptor.[67,68]

Upon irradiation of an organic PV, an exciton is created in the absorbing phase, which is followed by a very rapid electron transfer (<200 fs)[69] to the C_{60}/PCBM phase (photo-induced electron transfer). Since all the other known competing relaxation processes in

conjugated polymers occur on time scales that are orders of magnitude larger, this ultra-fast charge transfer must have a quantum efficiency of approximately unity, i.e., nearly all excitons that are created near the donor–acceptor (D–A) junction are transferred to the PCBM phase. Recently, power conversion efficiencies of upto 5% were reported for mixtures of P3HT and PCBM solution-processed from chloroform and using various processing tricks.[70] Furthermore, organic tandem and multi-junction photovoltaic cells are pursued to increase efficiencies.[71]

The fundamental importance of the field of conducting polymers and their great promises as enablers of a new electronic technology were recognized by awarding the 2000 Nobel Prize in Chemistry to their discoverers: Alan J. Heeger, Alan G. MacDiarmid, and Hideki Shirakawa.[72]

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