

Electronic transport properties aspects and structure of polymer-fullerene based organic semiconductors for photovoltaic devices

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Available online 7 February 2006

Abstract

A series of polystyrene (PS) and fullerene (C₆₀) based thin films containing from 23 to 60 wt.% in fullerene were investigated. Initially, the films were characterised by Fourier Transform Infrared Spectroscopy (FTIR) spectroscopy where the characteristic absorption bands of both the fullerene and the polystyrene were revealed. The additional characteristic absorption bands due the grafted fullerene to polystyrene were revealed as well. The relative peak intensities provided with qualitative information of the films stoichiometry in terms of the fullerene's amount that was grafted to polystyrene. The optical properties of the films were investigated by spectroscopic ellipsometry (SE). It was found that the increase of the fullerene's amount that was grafted to polystyrene results in an increase of the absorption coefficient α , refractive index n , extinction coefficient k as well as in the dielectric constant ϵ_{∞} within the range between 2.4 and 2.8 for the lower and higher fullerene content, respectively. The films' $J-V$ characteristics, of the space charge limited current (SCLC) behaviour, showed increased currents with increasing the fullerene's content. The electron mobility was extracted and found to increase with increasing the fullerene amount, from 4×10^{-9} cm²/V s to 2×10^{-7} cm²/V s.

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Keywords: Fullerene derivatives; Spectroscopic ellipsometry; FTIR; Space charge limited current

1. Introduction

Indeed, photovoltaics is the only portable and renewable energy source available today. This gives boost to the continued research in this technology although classical photovoltaics are of an unfavourable cost structure compared with other renewable energy technologies due to the high cost investments into semiconducting processes technology [1]. The latter cost standpoint inspires the research of the solution processable organic semiconductors-based solar cells which are flexible and can be manufactured easily by printing and coating techniques. Additionally, organic semiconductors possess the unique advantage of the flexible chemical tailoring of their properties.

In such bulk heterojunctions organic solar cells, two different organic materials possessing electron donor and acceptor properties respectively are getting mixed to create a

material which is capable for generating charges under illumination, transporting and collecting these photogenerated charges to an external circuit.

Generally, a promising approach consists of the blend of strongly light absorbing conjugated semiconducting polymer and a soluble fullerene derivative deposited on a transparent conductive substrate by a simple solution processing technique [2,3].

A wide class of these conjugated polymers and oligomers [2,3] shows a subpicosecond photoinduced electron transfer from the excited state of the conjugated polymer into C₆₀ [4] which is a good electron acceptor capable of accepting as many as six electrons [5].

An example of such bulk heterojunction solar cells is the one based on blends of poly{[2-methoxy-5-[(3,7-dimethyloctyl)oxy]phenylene]vinylene} (MDMO-PPV) and 1-[3-(methoxycarbonyl)propyl]-1-phenyl-(6,6)-C₆₁ (PCBM) which has been recently fabricated and extensively studied with power conversion efficiencies up to 3% [6].

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One of the most important parameters that control the efficiency of a solar cell is the carriers' mobility of both the donor and the acceptor. It was reported from space charge limited current measurements that the electron mobility of pristine PCBM thin films (μ_e , 2×10^{-3} cm²/V s) is several orders of magnitude higher than the hole mobility of pristine MDMO-PPV thin films (μ_h , 5×10^{-7} cm²/V s).

The present study investigates the structure and electrical transport properties of the fullerene–polystyrene structures as a function of the amount of the fullerene that is grafted to polystyrene. This study aims to the performance optimisation of the PS-C₆₀ organic semiconductor as a function of the C₆₀ content. The PS-C₆₀ organic semiconductor is being used as an electron acceptor in DEH-PPV-PS-C₆₀ poly(2,5-bis(2'-ethylhexyl)-1,4-phenylene vinylene)) block copolymer [8], whose photovoltaic properties are under investigation in our laboratories.

2. Experimental

The physical properties of buckminsterfullerene (C₆₀) have been extensively investigated, and a variety of interesting conducting [9], magnetic [10], photochemical [11], and electrical properties [12] have been observed. Unfortunately the ability to fabricate devices based on C₆₀ has been limited due to its poor processability. To overcome this difficulty, a number of groups [13–15] have prepared polymeric derivatives of C₆₀.

Fullerene-containing polymers that are under investigation include systems where the polymer and fullerene are linked by one or several covalent bonds, as well as composites where bonding between the polymer matrix and fullerene is due to donor–acceptor and/or hydrophilic–hydrophobic interactions.

2.1. Material synthesis

Polystyrene (PS) is one of the polymers that have been successfully modified by fullerene [13,14,16]. Conventionally, fullerene-containing PS is made by the free-radical polymerisation of styrene with fullerene present either in the block or in an aromatic solvent. Another way of producing fullerene-containing PS that has been extensively discussed in the literature is anionic polymerisation involving the formation of active fullerene-containing centres. With this method, complex stellate and dumbbell-like structures have been produced [15,18].

The C₆₀ containing PS materials that were used in the present study were prepared by the cycloaddition reaction [19] of azido-substituted polystyrenes with C₆₀. The ease of preparation of azide-containing polymers, the lack of cross-linking, and the retention of the fullerene's electronic properties make this procedure a simple and versatile method for the synthesis of fullerene-containing polymers.

The required azidomethyl-substituted polystyrenes were prepared as shown in Fig. 1, with the percentage of azidomethyl units being controlled by the initial feed ratio. Although the chemical reaction itself is outside the scope of the present study, some more important information that concerns

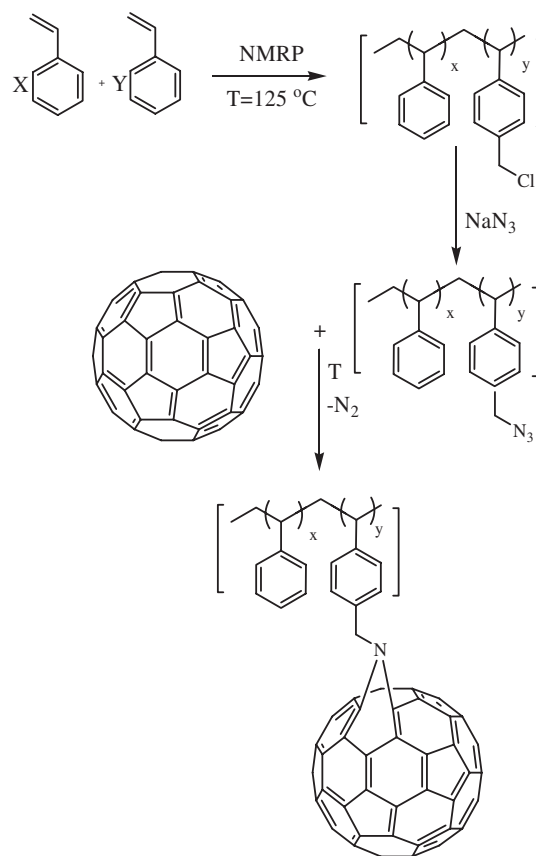


Fig. 1. Schematic of the reaction of C₆₀ with azido-functional polystyrene.

the structure of the resulting material will be given later in the FTIR section. More precise information about the technique itself can be found in [16,17,19].

2.2. Thin film deposition

The ITO covered polyethylene-terephthalate (PET) substrates were initially fluxed with ethyl alcohol and acetone, cleaned by ultrasonic treatment in isopropanol and acetone mixtures and finally by UV ozone treatment. Subsequently a layer of poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS) was spin-coated from an aqueous dispersion under ambient conditions onto the ITO substrates and the layer dried by thermal annealing. The active layers (PS-C₆₀) were spin-coated on the PEDOT:PSS layer in nitrogen atmosphere from 1,2-dichlorobenzene solutions. Prior to spin coating the solutions were filtered with a 0.2 μm PTFE filter in order to eliminate the incorporation of any aggregates within the films. Films of typical thicknesses (measured independently by spectroscopic ellipsometry and profilometry) at the range from 50 to 100 nm were deposited. Initially, the films were stored in high vacuum (2×10^{-6} mbar) for 4 h and for 24 h in the glove box nitrogen atmosphere for eliminating the presence of the solvent. Finally, a LiF layer of a thickness of 1 nm and 40 nm of Al contacts were deposited by thermal evaporation under high vacuum. More films for each C₆₀ amount were deposited on low-doped Silicon substrates for FTIR and spectroscopic ellipsometry measurements.

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