



Charge transport in thin films of MDMO PPV dispersed with lead sulfide nanoparticles



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ABSTRACT

Enhanced hole mobility is observed in thin films of poly(2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene) (MDMO PPV) dispersed with Lead Sulfide nanoparticles. Changes in the polymer film morphology upon the dispersion of nanoparticles and its influence on the enhancement of carrier mobility is investigated using Photoluminescence (PL) and Raman spectroscopic studies. PL and Raman studies show that at low concentrations of nanoparticles there is an enhancement in polymer interchain interaction. At higher concentrations, the interchain interaction decreases. Raman studies show that the observed enhancement of polymer interchain interaction at low nanoparticle concentration is due to the tighter packing of polymer chains. Thus, at low nanoparticle concentration, the observed increase in mobility is assigned to both the enhancement in interchain interaction and tunneling of carriers through nanoparticles. At higher nanoparticle concentrations the observed enhancement of mobility can be totally attributed to the tunneling of carriers through nanoparticles.

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1. Introduction

Realization of the importance of solar energy to meet the future energy demands and the concurrent rapid growth of photovoltaic market has fueled the efforts to harness solar energy in a cost effective manner [1]. Among the various available technologies, organic solar cells are considered as one of the best cost effective alternative to the conventional inorganic solar cells [2–5]. This understanding led to the unimpeded growth in the research to gain better insights to the device functioning and develop organic solar cells with higher efficiency [2–5]. Many combinations of materials, such as polymer–small molecule [3,4], polymer–polymer [6,7], small molecule–small molecule [8,9], polymer–inorganic nanomaterials [10–13] etc., are used for organic photovoltaic applications. Among these, the polymer–inorganic nanomaterials stand apart because of the certain distinct advantages with the use of inorganic nanomaterials such as nanoparticles, nanowires, carbon nanotubes etc. [10–14]. The use of inorganic nanomaterials provides the versatility to tune the band gap and thereby effectively utilizing the entire solar spectrum [10–14]. Other factors such as high electron mobility of inorganic materials for

better charge transport, large surface to volume ratio and low exciton binding energy for efficient free carrier generation are also advantageous for photovoltaic applications. In addition, these nanomaterials can be synthesized in solution using colloidal technique, where a good control of size and shape of the nanomaterials can be achieved [10–14]. The ability to synthesize nanomaterials in solution form also makes them compatible with various cost effective solution processing techniques suitable for large area device fabrication.

Variety of nanoparticles such as Cadmium Telluride (CdTe), Cadmium Selenide (CdSe), Lead Sulphide (PbS), Lead Selenide (PbSe), Zin Oxide (ZnO) [11–14] etc. are used as electron acceptors in polymer–nanoparticle (polymer nanocomposites) based solar cells. Even though polymer nanocomposites are promising for the development of cost effective and large area solar cells the understanding of the various optoelectronic properties of this class of materials is limited [15] and better insight to their properties is indispensable for fabricating efficient devices. An important factor that influences the various optoelectronic properties of active layers of polymer based solar cell is the active layer film morphology. Film morphology is highly dependent upon the type of nanoparticle, size of nanoparticle, concentration of nanoparticle, nature of polymer, processing conditions, solvent used for film casting etc. [16–19]. Tailoring the film morphology of the active layer is important to achieve efficient free charge carrier

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generation and transport of generated free carriers to the respective electrodes without significant recombination or loss due to traps. Balanced charge transport of electron and hole across the active layer is indispensable for attaining higher power conversion efficiency [20]. Therefore, charge transport is an important property that governs the performance of solar cells and it is highly influenced by the active layer film morphology. One of the interesting observation associated with the charge transport in polymer nanocomposites is the enhancement of the hole mobility upon dispersing the nanoparticles in polymer matrix, i.e. mobility increases with increase of nanoparticle concentration [21–24]. This was explained due to the tunneling of charge carriers through nanoparticles that leads to the enhancement of the mobility [22–24]. Addition of nanoparticles to the polymer matrix may alter the polymer film morphology and have a role in the enhancement of mobility. This point was not investigated well in earlier reports. In this study, we investigate the variation in carrier mobility in poly(2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene) (MDMO PPV) dispersed with PbS nanoparticles (MDMO PPV:PbS) using time of flight transient photoconductivity. MDMO PPV is a model conjugated polymer, which was widely studied for organic-inorganic hybrid photovoltaic applications [25]. Fig. 1a shows the chemical structure of the MDMO PPV. PbS nanoparticles, which possess interesting properties such as narrow band gap, large exciton Bohr radius and strong quantum-size effect, also been investigated extensively for developing various devices such as organic inorganic hybrid solar cells, infrared detectors, electroluminescence devices etc. [26]. Photoluminescence (PL) and Raman spectroscopic studies on thin films of MDMO PPV:PbS are carried out to understand the changes in polymer film morphology upon incorporation of nanoparticles. Our studies show that at low concentration of nanoparticles the enhancement of mobility upon addition of nanoparticles can be attributed not only to the tunneling of carriers through nanoparticles but also to the enhancement in polymer interchain interaction. At higher concentration, the enhancement of mobility can be totally attributed to the tunneling of carriers through nanoparticles, as suggested by earlier reports [22–24].

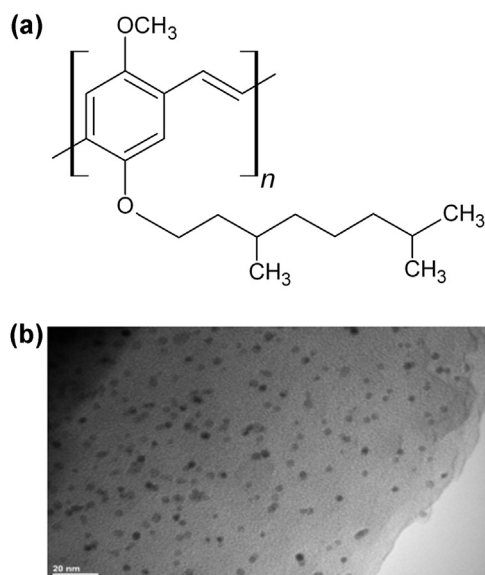


Fig. 1. (a) Chemical structure of MDMO PPV (b) TEM image of PbS nanoparticles dispersed in MDMO PPV thin film.

2. Details of nanoparticle synthesis and experimental techniques

2.1. Synthesis of PbS nanoparticles

PbS nanoparticles are synthesized by following the procedure reported elsewhere [27]. Lead Acetate solution (~ 1 mM) is prepared in a solvent mixture of Toluene (~ 25 ml) and DMSO (~ 6 ml). To this solution ~ 0.5 ml Oleic acid, which acts as a capping agent, is added and the mixture is kept for stirring for few hours to obtain a homogeneous bright yellow colored solution. Na_2S solution (~ 2 mM) in Toluene, which is the source of sulfur, is added drop by drop into this homogeneous lead precursor solution under constant stirring. Upon addition of the Na_2S solution, the bright yellow colored solution immediately turns to reddish brown and finally to dark brown, which indicates the formation of nanoparticles. Using methanol/ethanol, the synthesized nanoparticles are precipitated, extracted and then re-dispersed in Toluene. The precipitation and re-dispersion of nanoparticles are performed for four times and the final extracted nanoparticles are dried into powder form. The synthesized nanoparticles are spherical in shape with size of ~ 3 – 5 nm. The synthesized nanoparticles are mixed with polymer at required proportion by weight in a common solvent. Fig. (1b) shows the Transmission electron images (TEM) of PbS nanoparticles dispersed in MDMO PPV.

2.2. Sample preparation

Solutions of MDMO PPV:PbS, dispersed with nanoparticles at various proportions by weight (2%, 5%, 7% and 10%), are prepared using Chlorobenzene as solvent. Thin films using these solutions are prepared by drop casting on to neatly cleaned Fluorine doped Tin Oxide (FTO) coated glass. Films are allowed to dry in vapors of same solvent. Residual solvent from these thin films are removed by keeping them in vacuum oven for 12 h at 50°C . The sample geometry FTO/MDMO PPV:PbS/Al is completed by thermally depositing a semi-transparent aluminum (Al) top electrode (~ 50 nm) at a base pressure of 10^{-6} mbar. This allows excitation of the sample either through FTO or top semi-transparent Al electrodes. Thicknesses of the fabricated devices are of few microns, measured by Alpha-Tencor surface profiler. Capacitance and device area of the fabricated devices are ~ 10 pF and ~ 4 mm² respectively. In these samples, field dependence of hole mobility is obtained using Time of Flight (TOF) transient photoconductivity technique.

2.3. Time of flight (TOF) transient photoconductivity

Field dependence of hole mobility in these samples, at room temperature ($T = 300$ K), is determined using conventional small signal Time of Flight (TOF) transient photoconductivity technique [28]. A variable DC potential is applied across the device in reverse bias such that charge injection from the electrodes can be neglected, i.e. Al biased positive. A 5 ns laser pulse, at $\lambda = 490$ nm, from EKSPLA-NT342 laser is used for generating a thin sheet of charge carriers near to Al/MDMO PPV:PbS interface. Laser intensity is adjusted using optical density filters so that total charge generated is $\ll 0.05CV$, where C is the capacitance of the device and V is the voltage applied across the device. The time resolved photocurrent is recorded as a voltage across a load resistance using an oscilloscope. Analysis is carried out on the current pulses which are averaged over several laser pulses. The carrier transit time (τ) across the sample is obtained from photocurrent pulse and the mobility is calculated using $\mu = L^2/V\tau$, where L is the thickness of the sample. Both dispersive and non-

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