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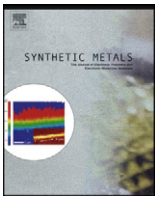

Confinement-induced enhancement of hole mobility in MEH-PPV

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ABSTRACT

Template wetting nanofabrication was used to create high-aspect-ratio, nanotubular structures from the semiconducting polymer poly(2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylenevinylene). Carrier transport in these nanostructures was determined to be space-charge-limited and thermally activated, with low-field mobilities shown to up to three be orders of magnitude higher than those typically reported for thin-films of the same material. Ultraviolet–visible spectroscopy showed a 0.3–0.6 eV reduction in the leading-edge absorption energy of polymer nanotubular structures compared to thin-films or solutions. Photovoltaic devices constructed from the nanotubes demonstrated photovoltaic fill factors superior to those measured in similarly constructed thin-film devices. These results are indicative of a confinement-induced ordering present in the nanotubular material.

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1. Introduction

Semiconducting and optoelectronic π-conjugated polymers have been studied for a wide range of electronic device applications, most notably light-emitting diodes [1,2], photovoltaics [3,4], and thin-film, field-effect transistors [5–7]. Much interest surrounding these materials has been generated due to the ability to inexpensively process and fabricate devices from them using solution based methods at ambient temperatures with techniques such as inkjet printing, spin coating, and microstamping [8,9]. However, several key obstacles including very limited exciton lifetimes and poor charge carrier mobilities, due in part to the lack of molecular order in most thin-films, must be addressed in order to enhance device performance metrics [10].

With the exception of a few small molecules and various polythiophenes, carrier mobilities above $10^{-5}\,\mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ are rarely observed in organic semiconductors [11,12]. Carrier transport between adjacent organic molecules, which is recognized to occur via hopping mechanisms, is primarily responsible for experimentally observed limitations of carrier mobility [10,11]. The high degree of molecular ordering found in materials such as pentacene [13] and self-organizing poly(3-hexylthiophene) (P3HT) leads to greater π–π orbital overlapping, enhancing intermolecular transport and thus charge carrier mobilities [11]. Replication of such ordering in field-effect transistors fabricated from the typically disordered polyfluorene copolymer, F8T2, not only demonstrated that ordering improved carrier transport, but also that mobilities measured when conduction was parallel to the length axis of the molecule were orders of magnitude higher than those perpendicular [14]. This confirmed interchain transport processes to be the limiting factor for F8T2, as was previously shown for P3HT [11]. Aside from the use of substrates that have been coated with alignment layers which act to orient the polymer chains (as in ref. [14]), several other methods have been employed to improve molecular ordering. The most notable efforts relevant to this work include production of nanoscale fibers and tubules from various polymers using techniques such as electrospinning [15], nanofluidic-lithographic methods [16], template synthesis [17], and template wetting [18].

Developing a technique known as template synthesis, Martin demonstrated multiple order of magnitude increases in conductivity in various conducting polymers when chemically synthesized within nanoporous membranes [17]. These increases were attributed to directional orientation of the molecules such that their length axes were parallel to both the length of the fiber and the direction of externally applied electric fields [19]. Later work by Steinhart et al. demonstrated a different process, template wetting nanofabrication, to be a simple and effective alternative to

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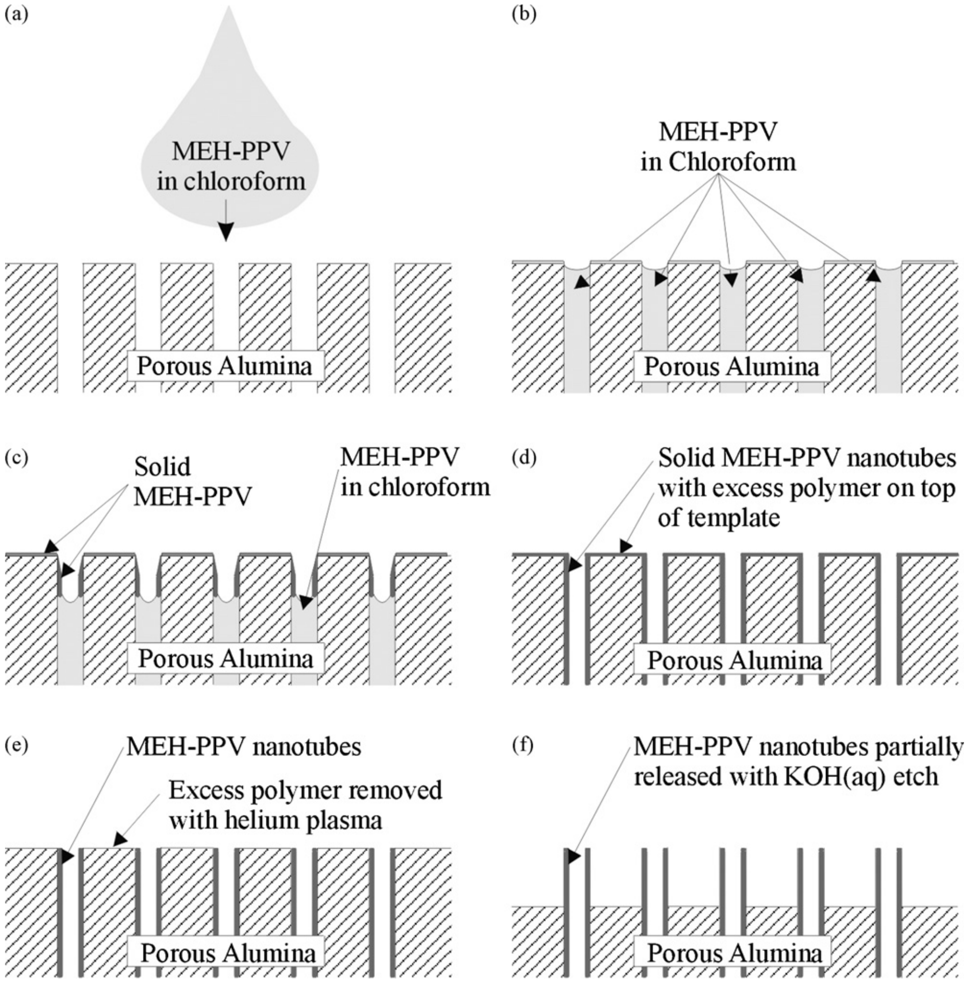

Fig. 1. Template wetting nanofabrication of MEH-PPV nanotubes. An MEH-PPV solution is applied to a porous alumina template (a) and surface tension draws the solution into the pores (b) evaporation of the solvent (c) leaves behind a coating of polymer on the pore walls (d). Excess polymer on the top of the template (d) is removed with a helium plasma (e) and if desired, the nanotubes are partially released by selectively etching the alumina with a KOH solution (f).

template synthesis for the creation of such polymer nanostructures [20]. The process utilizes wetting phenomena to create uniform coatings of low surface energy materials, such as many polymer solutions and melts, on porous template materials with high surface energy such as alumina. Nanotubes and nanowires created in this manner have shown enhanced characteristics as compared to bulk materials, including curvature induced molecular order and crystallinity of polymers within the nanopore [21].

Low- or zero-field hole mobilities in the widely studied, amorphous, photoactive polymer poly(2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) are typically observed to be on the order of 10^{-6} cm^2 V^{-1} s^{-1}, whether measured by time-of-flight techniques [22] or current density analyses [23]. In the present work, template wetting nanofabrication is used to create MEH-PPV nanotubes with a high degree of molecular order. Optoelectronic properties in these MEH-PPV nanostructures were investigated by examination of temperature dependent, space-charge-limited (SCL) currents, simple photovoltaic devices, and by means of ultraviolet–visible (UV–vis) spectroscopy.

2. Experimental

2.1. Materials

Anodic porous alumina membranes (Whatman Anodisc) were used as templates for nanofabrication. These membranes were 60 μm thick with either 100 nm or 200 nm nominal pore diameters and pore densities of 10^{10} and 10^9 pores per cm^2, respectively. MEH-PPV (MW 70,000–100,000, as powder) and poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS, 1.3% dispersion in H_2O) were obtained from Sigma–Aldrich. MEH-PPV wetting solutions were created by dissolving the polymer in chloroform (2 mg/mL).

2.2. Sample preparation

The template wetting nanofabrication process is illustrated in Fig. 1. Porous alumina templates were first wetted with the MEH-PPV wetting solution. The bulk density of the polymer and template porosity were used in conjunction with the assumption of cylindrical pores to calculate the mass of polymer required to make nanotubes with wall thickness of 5 nm. Surface tension effects cause the polymer solution to wet the alumina template and penetrate into the nanoscale pores. Samples were left in a fume hood for several hours to allow complete evaporation of the solvent to occur, leaving behind a solid polymer film coating the pore walls of the template. Residual polymer on the outer template surfaces was removed by exposing the substrate surfaces to helium plasma at 200 mTorr. Given the low temperature and short time of the etching process, along with the high-aspect-ratio of the pores, the plasma has no significant effect on the polymer embedded within the template [24]. In order to enable imaging of the resulting poly-

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