

Controlled growth of dibenzotetraphenylperiflanthene thin films by varying substrate temperature for photovoltaic applications

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ABSTRACT

We investigate the growth of a novel organic photovoltaic material, dibenzotetraphenylperiflanthene (DBP) thin film by ultra-high-vacuum evaporation. It is shown that the surface morphology and crystalline order of DBP thin films are greatly changed by growth temperature. Especially, at 90 °C, the DBP exhibits a unique orientated growth with long well-directional grain structures. The power conversion efficiency of the device using DBP grown at 90 °C is enhanced by a factor of 61% relative to that of the device using DBP grown at room temperature due to the improvements in photocurrent, open-circuit voltage and fill factor. The results indicate the feasibility of the controlled growth of DBP thin films, which plays a crucial role in improving the performances of photovoltaic devices.

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1. Introduction

Organic photovoltaic (OPV) devices based on small molecules and polymer have been attracting great interests as a potentially useful source of renewable energy due to their important advantages including low weight, low-cost production and flexibility [1]. The processing methods generally used for small molecule (vacuum evaporation) and polymer (solution process) materials govern the structure of OPV using these materials. Polymer OPVs using bulk heterojunction exhibit more promising efficiencies because high-quality blend layer can be obtained via solution process. Over 7% power conversion efficiencies (PCEs) have been reported in polymer OPV [2]. Especially, solution process has been well developed based on industrial considerations. The solution process can be environmental friendly, and it is also feasible for large-scale applications and all-layers preparation including the metal electrode [3–7]. On the other hand, significant progresses

have been achieved since first efficient donor/acceptor (D/A) planar heterojunction device by vacuum evaporation was reported, and the PCE of small-molecule OPV devices reach over 5% [8,9]. Vacuum evaporation of small molecule enables better control on the thickness, crystalline structure, morphology and large-scale uniformity of organic thin films. Thus, vacuum deposition is also an indispensable technique for developing future advanced organic devices with desired structures, especially for multi-layered structures in the nanometer range [10].

The biggest challenge of OPV is that PCEs of these devices using various materials and device structures are still not sufficient for practical applications. Generally, PCE is limited by several instinct factors including light absorption, exciton diffusion, charge collection and charge transfer efficiencies [11]. Developing new organic materials with optimized molecular ordering and morphology is becoming crucial for further improving OPV performances. Among various materials, dibenzotetraphenylperiflanthene (DBP) [12] is regarded as one of promising donor materials due to the promising electronic structure with lowest unoccupied molecule orbit (LUMO) energy (3.5 eV) and highest occupied molecule orbit (HOMO) energy (5.5 eV), especially the high light absorption coefficient. DBP has relatively high thermal stability and suitable sublimation temperature (350–450 °C), but limited solubility in many organic liquids. Thus, evaporation is the best choice for preparing DBP film. Very recently, a high PCE of 3.6% is reported in a simple planar

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heterojunction device using DBP as a donor and fullerene (C60) as an acceptor, respectively [13]. However, detailed discussion based on the growth of the DBP thin films for photovoltaic application is still scarce. Furthermore, it remains challenge to find out suitable process in order to better control the molecular ordering and film morphology of organic materials for device application [14]. In this paper, we investigate the growth of evaporated DBP thin films by varying the substrate temperature from room temperature to 150 °C in an ultra-high-vacuum (UHV) system. Significant variations in the surface morphology are observed in the samples grown at and above 90 °C. Planar DBP/C60 heterojunction devices are fabricated, and the effects of the structure and morphology of DBP thin films on photovoltaic properties are discussed in detail.

2. Experimental

Commercial pre-patterned indium-tin-oxide (ITO) coated glass was used as substrate. Prior to deposition, the substrate was treated by oxygen plasma for 30 mins. DBP (e-Ray Optoelectronics Technology) and sublimated C60 (Frontier Carbon Corporation) were used without further purification. The thickness and growth rate were monitored by a quartz-crystal oscillation and verified by a surface profiler (Veeco, Dektak 8). 20 ± 2 nm DBP thin films were prepared by an UHV evaporation system at a deposition pressure of around 2×10^{-6} Pa. The growth rate was kept at 0.05 ± 0.01 Å/s by controlling the Knudsen-cell temperature. The substrate temperatures were adjusted from room temperature (25 ± 1 °C) to 150 °C. To avoid the structural variations in C60 films, after deposition of DBP films, the samples were transported to another vacuum system via a sealed container filled with N₂. C60 films and electrodes were evaporated on 4 different substrates at one time. 80 ± 5 nm C60 film was deposited at room temperature with a growth rate of 0.2 Å/s on the surface of DBP film to fabricate planar DBP/C60 heterojunction. 0.1 nm LiF and 100 nm Al layers were deposited as cathode. The typical cell area was 0.04 cm². The surface morphology was studied by atomic force microscopy (AFM, SII, E-sweep) with dynamic mode. The

crystallinity of DBP films was discussed by reflection high energy electron diffraction amplified with a micro-channel plate (MCP-RHEED). The current density versus voltage (*I*–*V*) characteristics of the devices were measured in the dark and under simulated AM 1.5G solar illumination by using a Keithley 2400 Digital Source Meter at room temperature. Incident power was adjusted using a calibrated silicon photodiode to match 1-sun intensity (100 mW/cm²). Incident photon-to-electron conversion efficiency (IPCE) spectra were collected using a xenon lamp, integrated with a computer controlled monochromator. Source power spectrum was measured using a calibrated silicon photodiode.

3. Results and discussion

The chemical structure of DBP is shown in the insert of Fig. 2. The effects of substrate temperature on the surface morphology of DBP films were investigated by AFM. Fig. 1 shows the top view AFM images ($1 \times 1 \mu\text{m}^2$ scanning square) and line profiles of DBP thin films. It is seen that the grain size and shape are strongly dependent on the substrate temperature. The micrographs of the samples prepared at and below 70 °C show smooth surfaces with similar morphology, which includes some large clusters composing of small round grains with diameter ranging from 20 to 40 nm. In contrast, significant variations can be seen in the samples prepared at and above 90 °C, as shown in Fig. 2. Especially, at 90 °C, the sample exhibits a series of stripe-like structures with well-directional grains, indicating a unique orientated growth of DBP film. At 110 °C, the stripe-like morphology disappears, which is replaced with relatively smaller grains with random shape. The grains seem to be arranged along a directional line. Moreover, further increasing temperature up to 150 °C leads to larger pillar-like grain structures. Film thickness of 20 ± 2 nm was confirmed by Dektak surface profiler, indicating the average layer thickness in a millimeter scale (usually above 1 mm). Note that the height of some of grains is even larger than the film thickness as shown in the line profile in Fig. 2(e). The results imply the formation of nano-sized discontinuous layer or

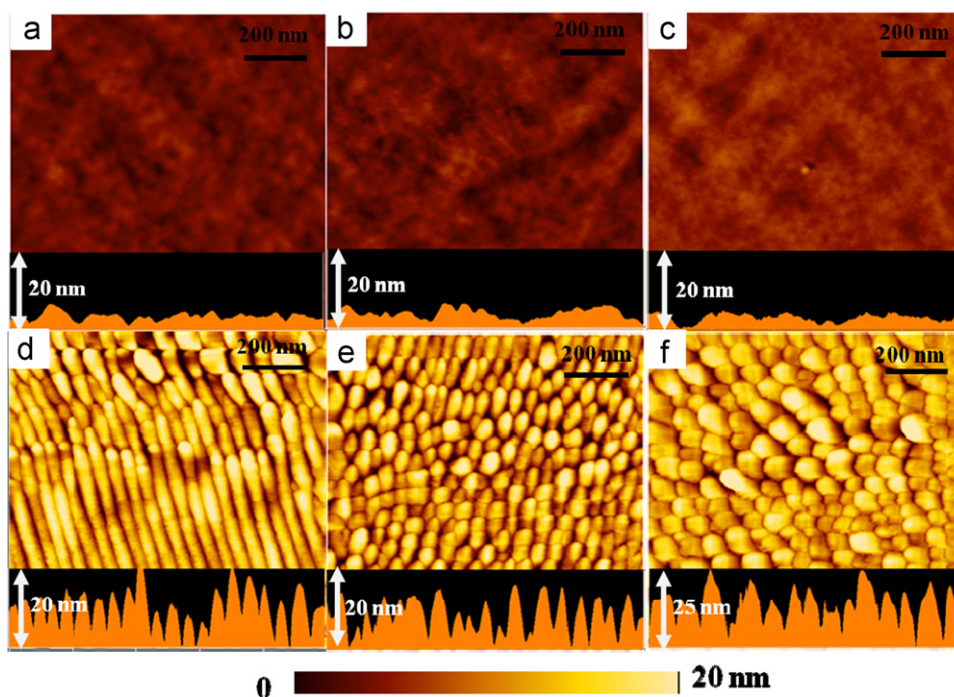


Fig. 1. AFM images of DBP thin films. (a) RT; (b) 50 °C; (c) 70 °C; (d) 90 °C; (e) 110 °C and (f) 150 °C. The contrasts are 0–20 nm for (a)–(e), 0–30 nm for (f).

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