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High-density organic photovoltaic modules: Mask-free fabrication using nozzle jet printing and oblique deposition

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ABSTRACT

We propose a cost-effective, mask-free fabrication process for high-density organic photovoltaic (OPV) modules based on oblique vacuum-deposition of small molecules over nozzle-jet printed microstructures. The contact angle of these microstructures is varied through surface modification, enabling them to function selectively either for separation or for passivation. An area utilization ratio as high as 95.6% with respect to a designated illumination area is demonstrated with the proposed method in series-connected OPV modules based on active layers of C_{70} doped with di-[4-(N,N-ditolyl-amino)-phenyl] cyclohexane (TAPC) molecules.

1. Introduction

Organic solar cells have gained attention as an alternative renewable energy source due to their potential for low-cost fabrication and unique features. Recently, the efficiency of organic photovoltaic (OPV) cells has reached 9–10%, which has long been regarded as an important milestone required to achieve before commercialization [1,2]. In order to realize their full commercial potential, a fabrication method for scale-up from a small single cell to a large module also needs to be secured so that they can fulfill the various power requirements demanded by an external load. Inorganic thin-film photovoltaic (PV) technology has taught us that a significant efficiency drop might occur during the integration of unit cells into large-scale modules [3,4]. This is mainly due to the limited sheet conductance ($G_{sh}^{(TC)}$) of transparent conductors (TCs), the optical shade effect due to an auxiliary metal grid used to supplement the limited $G_{sh}^{(TC)}$, and/or the presence of cell-to-cell interconnecting areas, which are optically inactive. The latter is unavoidable in realizing a series-connected module with a high voltage output, and thus an overall module design has to be carefully optimized with those interconnecting regions included. The challenge is that the interconnecting region cannot be made arbitrarily small due to the limited resolution of a fabrication technique used to form such regions. Things can easily get worse when it comes to OPV modules [5,6] mainly due to the limited processability of organic materials that prohibits use of high-resolution patterning techniques like photolithography or laser ablation that are commonly available for inorganic technologies. Furthermore, high-throughput processes are pursued in OPV technologies for low-cost fabrication, and they often accompany a significant compromise in patterning resolution, making high-density OPV modules even more challenging to realize [7–10]. In fact, inactive regions in most series-connected OPV modules have been as wide as several millimeters [10,11], an order-of-magnitude larger than that of inorganic thin-film PV modules adopting a laser ablation technique. Hence, importance of developing high-density module fabrication schemes compatible with organic materials cannot be overemphasized. In this work, we explore a new fabrication protocol that leads to high-density series-connected OPV modules with area utilization comparable to that of inorganic thin-film PV modules. With an emphasis on the importance of cost-effectiveness of OPVs, the proposed scheme is based on a mask-free deposition and tries to minimize use of costly processes like photolithography.

2. Experimental: module fabrication process and characterization

Fig. 1(a)–(c) shows the overall structure and circuit diagram of a series-connected OPV module and the fabrication process proposed in this study. First, bare ITO glass substrates (Eva SNP Co.) were patterned through standard photolithography process for a module scheme where the gaps between the ITO patterns were defined as 100 μm while the width of ITO for each unit cell was assigned as 0.64 cm. After the ITO patterning, an ultrasonic-based

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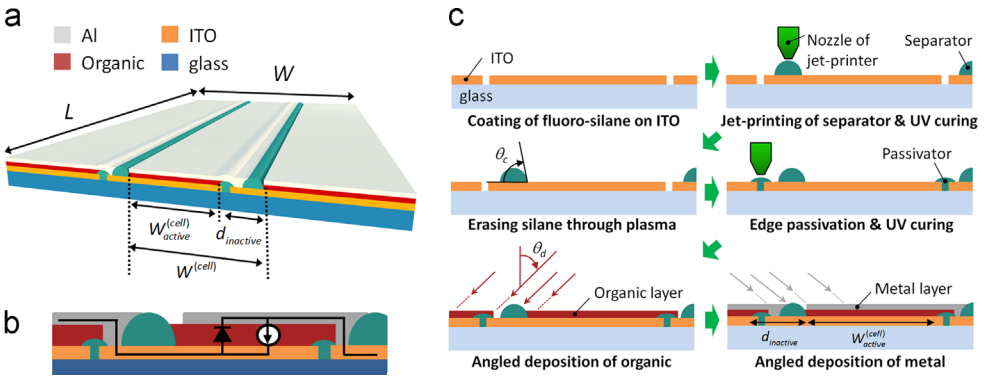

Fig. 1. (a) The schematic device structure and (b) the equivalent circuit diagram of our proposed module and (c) its fabrication process. In (a), ITO and Al correspond to an anode and a cathode, respectively. In (b), the direction of arrow in the current source symbol indicates the direction of photocurrent flow.

cleaning [12], and air plasma treatment (PDC-32G, Harrick plasma), the substrates were treated for hydrophobic surfaces with (tridecafluoro-1,1,2,2-tetrahydrooctyl) -1-trichlorosilane (TDF; United Chemical Technology) in a vacuum desiccator for 7 min. On top of these substrates, line-shaped microstructure arrays with a high contact angle (θ_c) were formed as 'separators' in a controlled manner using nozzle-jet printing of UV curable epoxy resin (Nagase, XNR5570). The nozzle jet printing was performed with an automatic dispenser system (Musashi Engineering Inc., ML-5000XII) having a desktop robot (Musashi Engineering Inc, Shot mini 100s). Air pressure up to 0.7 MPa and three-axis movement with a minimum operation step of 10 μm was obtainable with this system.

As can be seen from Fig. 1(c), the high-contact-angle linear structures work as an 'integrated shadow mask,' thereby defining the respective deposition areas for organic and metal layers while ensuring proper separation/interconnection for series-connected OPV modules. After curing the printed resin structures by exposing them under UV light (UVP, B 100AP, 100 Watt) for 10 min right after printing, the substrates were subject to air plasma to remove the previously coated fluorosilane and to make their surfaces hydrophilic. Then, the edges of the ITO patterns were 'passivated' by printing low-contact-angle structures with the same UV resin to suppress the leakage current that may be induced from an angled deposition of organic layers. The substrates were then brought into a vacuum thermal evaporation chamber (HS-1100, Digital Optics & Vacuum) for deposition of active layers and electrodes. The structure of the devices used in this study was glass/ITO (150 nm)/ MoO_3 (3 nm)/C_{70}:TAPC (5%, 60 nm)/Bphen (7 nm)/Al (80 nm), in which TAPC refers to di-[4-(N,N-ditolyl-amino)-phenyl] cyclohexane, and Bphen refers to bathophenanthroline [13]. MoO_3 (99.9995%, Alfa Aesar), C_{70} (99.0%, SES research), TAPC (Nichem) and Bphen (Nichem) were used as received, and evaporation of these materials were carried out at a base pressure of 1×10^{-6} Torr, at an oblique deposition angle (θ_d) of 25° with respect to the direction normal to the substrate. For the TAPC doped C_{70} layer, co-evaporation was carried out through controlling the deposition rate of two materials, which was set at 0.1 Å/s and 2.0 Å/s for TAPC and C_{70}, respectively. Before the deposition of the Al electrode, the deposition chamber was opened briefly exposed to a nitrogen environment in order to change the deposition angle to −25°. With the oblique deposition and the printed separator structures, the proposed OPV modules were fabricated without a shadow mask except for an auxiliary contact pad area. When the organic and the metal layers were deposited from different directions with the same angle, the printed linear separator arrays introduced shaded areas at each adjacent side of the separators as shown in Fig. 1(c). As a result, the interconnection between adjacent unit cells was well definable without any unwanted electrical connections with this mask-free deposition process.

Current density–voltage (J–V) characteristics were measured with a source-measure unit (Keithley 238) under AM 1.5G illumination (100 mW/cm^2) from a solar simulator (ABET technologies). The irradiance of the incident light was checked periodically using a calibrated Si photodiode which was cross-checked with a reference Si cell (Peccell Technologies Inc., PEC-SI01). The area of modules and cells was examined through a microscope having a digital camera.

3. Result and discussion

3.1. Formation of a linear microstructure by printing process

In the proposed module scheme, it is essential to print linear microstructures with contact angles (θ_c) of the structure defined in a controlled way. Their widths also have to be narrow enough in order to make the width of the inactive region ('d_{inactive}' in Fig. 1 (a)) as small as possible. The inverted pyramidal structure made by a photolithography based on a negative photoresist might be an ideal option for such well-defined micro-scale separators, but use of large-area photomasks would be too costly for OPV modules aiming at low-cost PV technologies and thus was avoided in this work.

At an early developmental stage, we initially tried printing photoresist (SU-8 2000.5) using an inkjet printer. As can be seen from Fig. 2(a) and (b); however, uniform linear structures were obtainable only on a hydrophilic surface with the low cross-sectional contact angle; on a hydrophobic surface, the droplets of SU-8 ejected from the nozzle of an inkjet tended to form another droplet instead of forming a linear structure. This behavior is regarded due to the low viscosity of the solution. The large surface energy difference between SU-8 and the hydrophobic surface promoted the drops to merge together without adhering to the substrate, making it almost impossible to build a continuous line. As an alternative, UV curable resin with a viscosity of 127 Pa·s, which is almost 12,700 times greater than SU-8, was adopted. Such a high viscosity material was printable with the aforementioned nozzle jet printer, which is commonly used for printing of edge-seal resin in encapsulation of organic devices.

Fig. 2(c) and (d) shows the scanning electron microscopy (SEM) images of the printed resin structures of the fabricated module covered with Al obliquely deposited at 25° with respect to the surface normal. For printing, a nozzle (Musashi Engineering Inc, SNA-30) with an inner diameter of 140 μm was used. The SEM images indicate that the proposed UV-curable resin can indeed form linear structures not only on a hydrophilic surface but also on