



Ultraviolet-ozone anode surface treatment and its effect on organic solar cells



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ARTICLE INFO

Article history:

Received 26 March 2016

Received in revised form 13 December 2016

Accepted 19 December 2016

Available online 21 December 2016

Keywords:

Organic solar cells

UV-ozone treatment

Indium tin oxide

Surface treatment

Repetitive illumination stress

Device degradation

ABSTRACT

The effect of ultraviolet (UV)-ozone-treated indium-tin oxide (ITO, anode) on the initial parameters and degradation in small-molecule organic solar cells (OSCs) with the structure of ITO/copper phthalocyanine (CuPc, donor)/fullerene (acceptor)/bathocuproine (cathode buffer)/Ag (cathode) was experimentally investigated. A UV-ozone exposure with a UV intensity of 13.5 mW/cm² on the ITO surface was examined. The results indicated that compared with OSCs with no UV exposure, the initial short-circuit current density and power-conversion efficiency η_p increased in the OSC by 20% and 13%, respectively, after 1 min of UV-ozone exposure. A repeated illumination stress comprising 3 s of illumination and 12 s of darkness was imposed on OSCs in air. The UV-ozone-treated OSCs showed excellent durability under the repeated illumination stress. After 10 min of UV-ozone exposure, the OSCs exhibited only an 8% decrease in η_p after 100 illumination cycles. However, with no UV exposure, η_p decreased by approximately 65%, and the OSCs developed strong S-shaped kinks in their current-voltage characteristics, suggesting an increase in series resistance at the ITO-CuPc interface. The UV-ozone treatment much reduces the carbon contaminants on the ITO surface and increases the clean surface with polar components of OH functional groups and negatively charged oxygen. This generates attractive force between ITO and CuPc film. The attractive force prevents further CuPc crystal disorder and void formation at the ITO/CuPc interface, and achieves the durability against the illumination stress.

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1. Introduction

Donor-acceptor organic solar cells (OSCs) were presented by Tang in 1986 and provided a new potential replacement for existing natural energy resources [1]. OSCs are promising for use as renewable energy sources because they have good mechanical flexibility, are made of low-cost materials, and are easy to fabricate. However, OSCs face limitations in terms of power-conversion efficiency η_p and stability under the illumination stress and air exposure required for practical use.

The improvement in η_p has been achieved, for example, by cathode buffer [2–5], anode buffer [6–12], and bulk-heterojunction [13,14]. The cathode buffer is placed between the organic acceptor layer and the metal cathode, acts as an “exciton-blocking layer” that prevents exciton quenching at the cathode/organic interface, leading to efficient exciton dissociation and eventually increasing η_p . The anode buffer is placed between the organic donor layer and the ITO anode, plays several roles in enhancing charge transport and improving the performance of OSCs. That is, the anode buffer effectively passivates surface defects, smoothes the rough ITO surface, modifies the molecular orientation and the crystalline structure of the donor, and adjusts the work function (WF) at the

anode/donor interface. At present, η_p as high as 9.2% for OSCs has been achieved [15].

However, obtaining long-term OSC performance remains a major task because OSCs deteriorate upon exposure to air and sunlight. In the degradation of OSCs, the current density-voltage (J - V) characteristics frequently deform from an exponential diode curve to an S-shaped kink curve in the positive bias region [16–20]. This change leads to a decrease in fill factor (FF) and η_p [11]. In the case of pentacene/fullerene (C_{60}) heterojunction OSCs, differences in the temporal changes of J - V characteristics were clearly observed among devices kept in air, under vacuum, and under vacuum with a UV filter [16]. The degradation phenomena were systematically investigated for small-molecule copper phthalocyanine (CuPc)/ C_{60} OSCs under illumination, air exposure, and electrical stress [21]. Understanding the degradation mechanism responsible for illumination stress is critical because OSCs generate power by accepting light energy.

Several solutions have been proposed to address these problems. The stability of OSCs under light irradiation is markedly enhanced by inserting a MoO_3 layer [22,23] or a set of ultra-thin Ag and thin pentacene layers [11,24] as an anode buffer layer. Other techniques such as device encapsulation [25] and the use of UV filters [26,27] retard the degradation process and maintain device stability, thereby enhancing device lifetime.

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Ultraviolet (UV)-ozone surface treatment is a useful technique for tuning the anode WF and optimizing the surface condition [28–33]. Numerous investigations have found that this treatment decreases surface roughness and eliminates organic contaminants. Applying UV-ozone treatment to indium-tin oxide (ITO) is expected to improve OSC performance by increasing the hole-extraction efficiency. In the field of organic light-emitting diodes (OLEDs), UV-ozone treatment leads to efficient hole injection by reducing the hole-injection barrier [34]. Because hole collection occurs at the anode in OSCs, modifying the interface between the organic layer and the anode should improve the device efficiency. However, it is unclear whether the same benefits obtained from UV-ozone surface treatment of OLEDs are obtainable in OSCs. Few reports have discussed the effects of UV-ozone treatment on OSCs, and the mechanism of how UV-ozone surface treatment affects charge transport in OSCs remains under debate. For example, Lo et al. reported that UV-ozone treatment potentially reduces device stability because it induces gap states that act as charge-recombination centers, which has implications for device lifetime [35]. Such reports are, however, limited. Thus, whether UV-ozone surface treatment degrades the stability of OSCs still remains unexplored.

In this work, we used ITO surface modification by UV-ozone treatment to improve device performance. The effect of UV-ozone treatment on the initial efficiency of OSCs is verified through a series of experiments. Device stability under illumination stress is further explored for devices subjected to UV-ozone treatment.

2. Experimental methods

Fig. 1 shows a schematic drawing of the structures of two types of OSCs: OSC0 and OSC1–OSC20. These devices are composed of ITO as the anode, a 20-nm-thick CuPc (Aldrich, 99%) layer as the donor, a 40-nm-thick C₆₀ (SES Research, 99.9%) layer as the acceptor, and a 10-nm-thick bathocuproine (BCP, Tokyo Kasei, refined product) layer as the cathode buffer layer. The OSC1–OSC20 samples were subjected to additional UV-ozone treatment of their ITO surfaces, where the sample numbers 0 through 20 in the device name indicate the UV exposure time in minutes. The organic materials were used as purchased.

Fig. 2 shows schematic structures of two types of hole-only devices with single 200-nm-thick CuPc layers sandwiched by ITO and Au/Ag metal electrodes, SL0 and SL10. Here the ITO surface of SL10 was subjected to an additional 10 min of UV-ozone treatment. These devices were designed to investigate the origin of the degradation due to

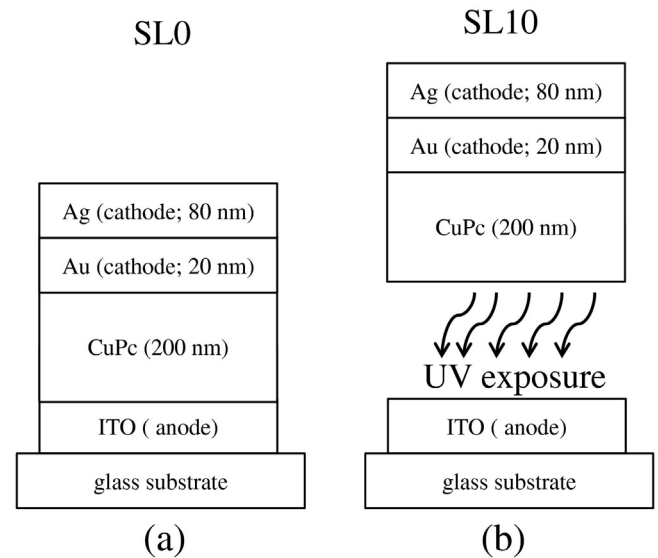


Fig. 2. Schematic structures of two types of hole-only devices with single 200-nm-thick CuPc layers sandwiched between ITO and Au/Ag compound metal electrodes. (a) SL0 without UV-ozone treatment on ITO surface. (b) SL10 with 10 min of UV-ozone treatment on ITO surface.

illumination of the OSCs shown in Fig. 1. The simple structures of these devices reduce the number of unnecessary interfaces in the OSCs [22], allowing us to focus on the changes in the bulk layers of CuPc and at their ITO–CuPc interfaces. The cathodes consisted of compound electrodes composed of 20-nm-thick Au layers and 80-nm-thick Ag layers. Here, CuPc was put in direct contact with the Au on the cathode side. The WF of ITO is 4.7 eV, and that of Au is 5.1 eV. The highest occupied molecular orbital (HOMO) of CuPc is 5.2 eV. Thus, in SL0 and SL10, the barriers against hole injection at their ITO/CuPc interfaces were higher than those at their Au/CuPc interfaces.

Fig. 3 shows a plan view of the device arrangement, with the numerical values in millimeters. Six devices were fabricated on ITO-precoated glass substrates (Mitsuru Optical Laboratory). The ITO was 1600 Å thick. After a broadband area with dimensions of 6 mm × 20 mm was masked,

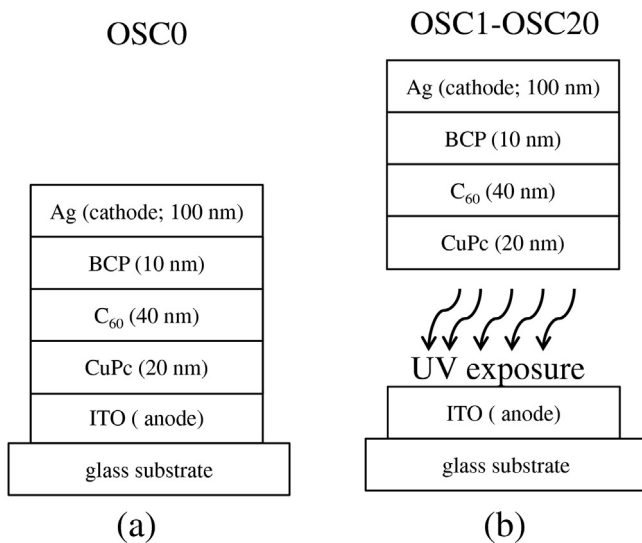


Fig. 1. Schematic structures of two types of OSCs with CuPc donor layers, C₆₀ acceptor layers, and BCP cathode buffer layers. (a) OSC0 without UV-ozone treatment on ITO surface. (b) OSC1–OSC20 with UV-ozone treatment on ITO surface.

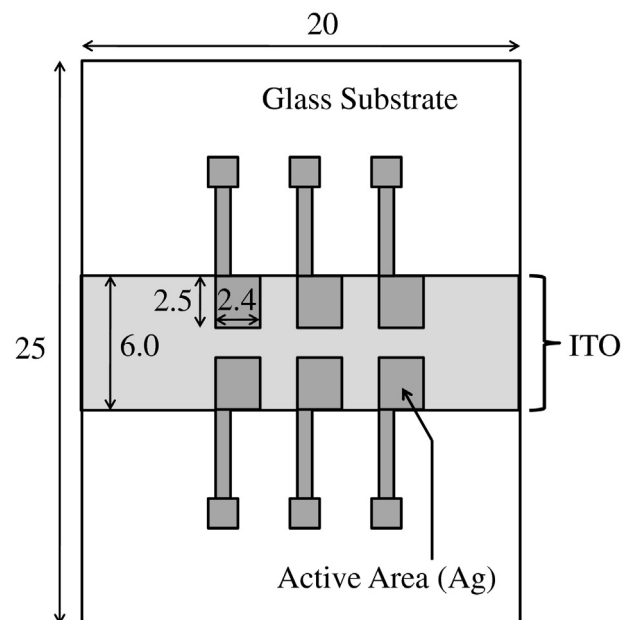


Fig. 3. Plan view of device arrangement on ITO substrate. Numerical values are in millimeters.

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