



Stability in organic solar cells with Ag/pentacene anode buffer under repetitive illumination

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ABSTRACT

The degradation phenomenon in small-molecule organic solar cells (OSCs) with a structure of indium-tin oxide (ITO, anode)/anode buffer/copper phthalocyanine (CuPc, donor)/fullerene (acceptor)/bathocuproine (cathode buffer)/Ag (cathode) was experimentally investigated. Two kinds of anode buffer layers, a 2-nm-thick pentacene layer and a 0.5-nm-thick Ag layer, were examined. The devices were subjected to repetitive illumination stress through the transparent ITO electrode in air. The ultra-thin Ag layer enhanced the ability of the devices to withstand the illumination stress. A thinner Ag layer resulted in better device performance. Resonant light absorption in the nanometer-scale Ag clusters probably accumulates trapped electrons near the ITO/CuPc interface, shrinks the depletion layer, enhances tunneling injection of holes, and suppresses the S-shaped kink during the illumination. The thin pentacene layer provides good ohmic contact between the anode and the CuPc layer, resulting in a larger short-circuit current. OSCs fabricated with both Ag and pentacene anode buffer layers exhibited only a 10% decrease in power conversion efficiency at 60 repetitions, whereas OSCs without an anode buffer layer exhibited a decrease of approximately 45%. The large decrease in efficiency was dominated by the decrease in the fill factor.

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1. Introduction

Organic solar cells (OSCs) have been recognized as a promising candidate for a renewable energy source because of their desirable properties such as good flexibility, low cost, large area, and easy fabrication. Tang made a major breakthrough in 1986 by introducing the concept of donor–acceptor (D/A) heterojunctions based on small molecular materials deposited via vacuum deposition [1]. The power conversion efficiency (η_p) of OSCs has been steadily improved through the use of cathode buffer layers [2–4], mixed-D/A heterojunctions [5,6], and new materials [7–9]. However, OSCs still suffer two major drawbacks: low η_p and a short lifetime.

Several degradation mechanisms that shorten the lifetime have been reported; these mechanisms include the diffusion of molecular oxygen, water, and metal atoms into the device; degradation of interfaces; degradation of the active material; interlayer and electrode atom diffusion; electrode reaction with organic materials; and morphological changes [10]. These mechanisms may be driven by illumination, external air exposure, temperature and/or electrical stresses [11]. The degradation of OSCs with a pentacene/fullerene (C_{60}) heterojunction was investigated in air with different relative humidities to clarify the effect of oxygen and water molecules [12].

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 [13–21]. This change leads to a decrease in fill factor (FF) and η_p [22]. Several theoretical models have been proposed to explain the mechanism of the S-shaped kink curve [19–23]; these models include corroded metal contact bringing about a thin current-limiting layer under forward bias voltage [20], charge accumulation induced by the cathode buffer layer [21], and charge blocking originating from the presence of strong interface dipoles [23].

Understanding the degradation mechanism responsible for illumination stress is critical because OSCs generate power by accepting light energy. The effect of illumination stress on the performance of OSCs has previously been evaluated. In the case of pentacene/ C_{60} heterojunction OSCs, an experimental study under constant solar illumination revealed two parallel mechanisms for degradation: a photo-oxidation process resulting in a decrease in photocurrent and an ultraviolet (UV) annealing effect reducing the fill factor [13]. 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. In addition, degradation in an OSC with an indium-tin oxide (ITO)/copper phthalocyanine (CuPc)/ C_{60} /bathocuproine (BCP)/Ag structure has been investigated under repetitive illumination stress in a high-vacuum chamber [11].

A promising method to improve the η_p and stability of OSCs under illumination is the introduction of a thin anode buffer layer of organic and inorganic materials between the anode and the donor layer. The organic anode buffer layer has been reported to play several roles in improving the performance of OSCs. First, the anode buffer modifies the molecular orientation and the crystalline structure of the donor, which enhances charge transport in the direction between the two electrodes

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[24–26]. Second, the anode buffer is efficient for adjusting the work function (WF) at the anode/donor interface, thereby reducing the potential barrier height of hole transport, achieving ohmic contact [27,28].

Perylene-3,4,9,10 tetracarboxylic dianhydride (PTCDA) has been investigated as a template layer for modification of the molecular orientation of metal-free phthalocyanine (H_2Pc) films [24]. A thin PTCDA layer deposited before CuPc growth templates the CuPc film structure, forcing the molecules to lie flat with respect to the substrate surface. This molecular orientation facilitates hole transport and improves hole collection [25]. However, PTCDA is an electron-transporting material, whereas an anode buffer should ideally be a hole-transporting material. Pentacene has also been proposed as an anode buffer material, because it is a hole-transporting material and exhibits a large hole mobility. The effect of a pentacene buffer layer on the molecular orientation of H_2Pc films has been investigated [26].

On the other hand, OSCs have been reported to exhibit a strong improvement of η_p when the transparent conductive anode is covered with an ultra-thin metallic film [29,30]. Appropriate matching between the WF of the anode and the highest occupied molecular orbital (HOMO) of the donor has been demonstrated to be the most important factor. Metal oxides have also been examined as an anode buffer layer in OSCs. The presence of thin metal oxides, MoO_3 , WO_3 and V_2O_5 , at the ITO–CuPc interface has been shown to substantially improve the performance of OSCs in terms of a reduction in the effective barrier against hole transfer from CuPc to ITO. [31–35].

The stability of OSCs under illumination is markedly enhanced by inserting a MoO_3 anode buffer layer [34,35]. In addition, the S-shaped kink under illumination is suppressed by inserting a set of ultra-thin Ag and thin pentacene layers between the anode and the donor organic layer [22]. In this case, a large hole-injection current is generated at the Ag–pentacene interface because of the tunneling effect when a positive external bias voltage is applied to the anode.

Several studies on the effect of anode buffer layers on the efficiency and stability of OSCs have been extensively investigated. However, the literature contains few systematic studies on the degradation under illumination in OSCs with combinational anode buffers consisting of organic and inorganic materials. Also, the physical mechanism has not been clarified.

In this study, we investigate the effect of illumination stress on the degradation of small-molecule OSCs with a structure of ITO/anode buffer/CuPc/ C_{60} /BCP/Ag. Two kinds of anode buffer layers, ultra-thin Ag and thin pentacene layers, are examined. The results show that the OSC with an ultra-thin Ag layer exhibits reduced device degradation and that the OSC with a thin pentacene layer exhibits an enhanced short-circuit current density J_{sc} , resulting in an increase in η_p .

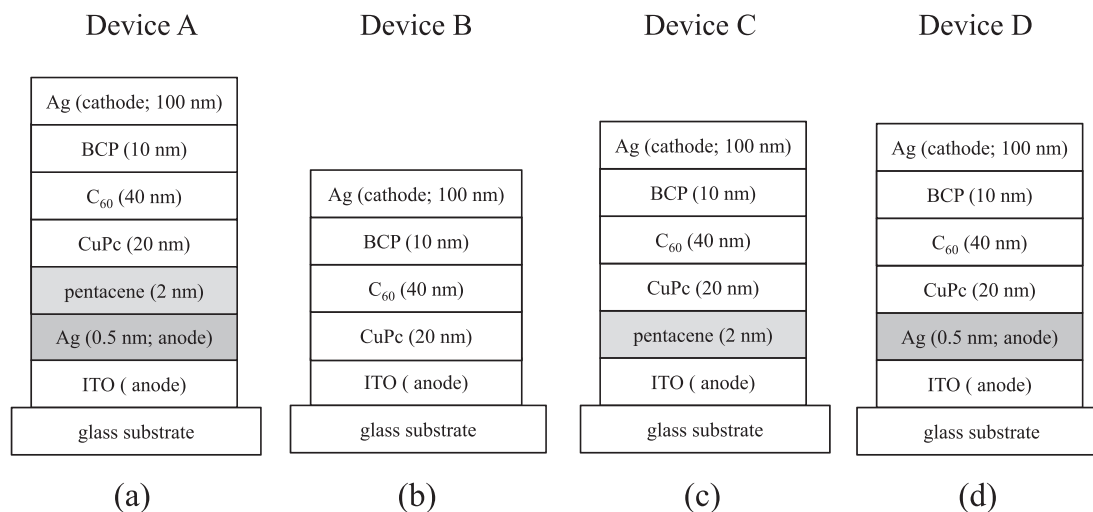
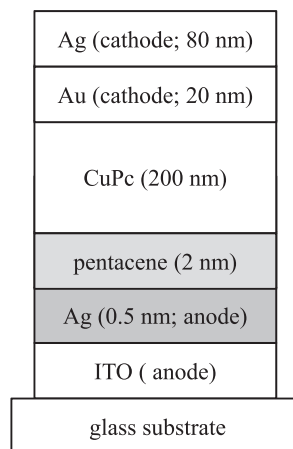


Fig. 1. Schematic structures of four kinds of organic solar cells: (a) Device A, with an anode buffer consisting of thin pentacene and ultra-thin Ag layers; (b) Device B, without an anode buffer (standard device); (c) Device C, with an anode buffer consisting of a thin pentacene layer; and (d) Device D, with an anode buffer consisting of an ultra-thin Ag layer.

Device E



Device F

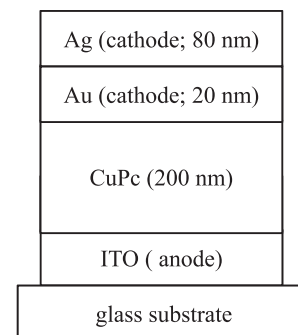


Fig. 2. Schematic structures of two kinds of hole-only devices with a single CuPc donor layer sandwiched between different metal electrodes; (a) Device E, with an anode buffer consisting of thin pentacene and ultra-thin Ag layers, and (b) Device F with no anode buffer.

2. Experimental methods

Fig. 1 shows schematic structures of four kinds of OSCs, Devices A–D. All four of these devices include a 20-nm-thick CuPc layer as the donor, a 40-nm-thick C_{60} layer (SES Research, 99.9%) as the acceptor, and a 10-nm-thick BCP layer (Tokyo Kasei, refined product) as the cathode buffer layer. Device A has an additional anode buffer composed of a 0.5-nm-thick Ag layer and a 2-nm-thick pentacene layer (Aldrich, 99%). Device C has a 2-nm-thick pentacene layer as an anode buffer. Device D has an anode buffer consisting of 0.5-nm-thick Ag layer. The CuPc (Aldrich, 90%) was purified three times by thermal gradient sublimation before use [36]. The other organic materials were used as-purchased. Device B, which has no anode buffer layer, was treated as a standard device.

Fig. 2 shows schematic structures of two hole-only devices with a single 200-nm-thick CuPc layer sandwiched by different metal electrodes, Devices E and F. These devices were designed to investigate the origin of illumination degradation in the OSCs shown in Fig. 1. The simple structure of these devices reduces the number of unnecessary interfaces of OSCs [34], allowing us to focus on the changes in the bulk layer of CuPc and ITO/CuPc or ITO/Ag/pentacene interfaces. A

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