



A surface modification layer capable of tolerating substrate contamination on transparent electrodes of organic electronic devices



Xun Tang, Min Qian, Dong-Ying Zhou, Lei Ding, Yun Hu, Liang-Sheng Liao*

Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Institute of Functional Nano & Soft Materials (FUNSOM), and Collaborative Innovation Center of Suzhou Nano Science and Technology, Soochow University, Suzhou, Jiangsu 215123, China

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ABSTRACT

An organic molecule, hexaazatriphenylene hexacarbonitrile (HAT-CN), is found that it can be used not only as a hole-injecting material but also a surface modification material to clean contaminated substrate electrodes for the fabrication of organic electronic devices. As an example, HAT-CN can modify or “clean” indium-tin-oxide (ITO) anode surface in organic light-emitting diodes (OLEDs). Negative effect from ITO surface contamination on the electroluminescence performance of OLEDs can be dramatically reduced with this modification layer. As a result, the OLEDs with the same device architecture but with different ITO surface conditions, even with intentional contamination, can all exhibit substantially identical and superior electroluminescence performance. The surface modification function of this material is feasibly useful for the real fabrications of OLEDs as well as for advanced research on other organic electronic devices.

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1. Introduction

Transparent electrodes are commonly used in organic electronic devices including organic light-emitting diodes (OLEDs), organic solar cells (OSCs), organic photodetectors (OPDs), and transparent organic field-effect transistors (OFETs), as well as in some inorganic electronic devices [1–3]. A transparent electrode usually functions as a substrate electrode, or a bottom electrode, on a surface of a transparent substrate, such as the electrode made from indium-tin oxide (ITO), indium-zinc oxide (IZO), and some other conductive metal oxides. The surface condition of the substrate electrodes is directly related to device performance. Any slight contamination on the surface of a substrate electrode will cause substantial change both on carrier injection behavior at the interface between the electrode and an adjacent organic layer and on operational lifetime of the device. For example, in OLEDs, the surface conditions of the ITO can cause substantial changes in drive voltage, current efficiency, and operational stability [4].

Therefore, in order to fabricate high quality organic electronic devices, a patterned substrate electrode on a transparent substrate must be thoroughly cleaned and modified before it could be used

for device fabrication. In a regular cleaning process, it involves the steps of mechanical scrub, fluidic wash, oven baking, and optical inspection. Following the cleaning process is the further treatment or modification on the clean electrode surface. There are several well-known surface treatment methods including plasma treatment (such as of oxygen plasma, C₂F₄ plasma, Cl₂ plasma, atmospheric plasma) [4–7], ultraviolet ozone treatment [8], chlorinated treatment [9], and wet treatment [10,11]. Only after the surface treatment, usually a modification layer (or interfacial layer) is then normally formed on the surface of the electrode, which may be a self-assembly monolayer [12,13], conducting layer (such as conducting polymer or thin metal layer) [14–16], semiconducting layer (such as CuPc, F₁₆CuPc, CuO_x, C₆₀, or MoO_x) [17–21], or one of thin insulating layers including SiO₂, Si₃N₄, metal oxides, Teflon, InCl₃, and LiF [22–27].

The purpose of these surface treatments is to control the cleanliness, morphology, and surface work function of the electrode. However, the work function at the electrode surface is very sensitive to the surface conditions. Even after the aforementioned surface treatments, surface contamination during substrate storage and substrate transfer is more or less inevitable prior to the deposition of an organic layer on top of the substrate electrode in a vacuum system [28]. This contamination causes a reduced surface work function and an increased carrier-injection barrier at the interface between the electrode and the adjacent organic layer,

* Corresponding author.

E-mail address: lsiao@suda.edu.cn (L.-S. Liao).

resulting in degraded and scattered device performance. From the manufacturing point of view, it is difficult to maintain an identical electrode surface condition on a daily fabrication base. Moreover, in the regular surface cleaning and treatment process, high volume of tap water, detergents, acetone, ethanol, and deionized water needs to be consumed. As a result, it not only causes waste of the resources, but also creates pollution to our environment. In addition, this is a time-consuming and energy-consuming process having negative influence on cost-effective production. So far, neither device manufacturers nor research groups would use as-prepared substrate electrodes to fabricate high-performance organic electronic devices without a thorough surface cleaning and treatment process. Therefore, it would be very useful if a reliable and cost-effective way could be found for producing high quality organic electronic devices with tolerated electrode surface conditions and with a simplified electrode surface modification process. In other words, if as-prepared substrate electrodes could be used directly for device fabrication, one could imagine how resources could be saved and how our environment could be protected to some extent. Here in this paper, we would like to take an ITO anode in OLED as an example to report that as-prepared substrate electrodes could be indeed used directly for device fabrication, which could even have better electroluminescent (EL) performance than that of the devices with traditionally cleaned ITO anode.

An organic molecule, 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN), has been reported as an n-type semiconducting material for use in organic electronics [29], as a hole-injecting layer on an oxygen plasma-treated ITO anode in OLEDs [30], and as a part of the intermediate connector in tandem OLEDs [31–33]. We further report here that HAT-CN can form a “cleaning” layer on different ITO surfaces. Negative effect from ITO surface contamination on the EL performance of OLEDs can be dramatically reduced with this “cleaning” layer. Therefore, the OLEDs with the same device architecture but with different ITO surface conditions can all exhibit substantially identical and superior EL performance.

2. Results and discussion

2.1. Effect of electrode surface conditions on EL performance

Shown in Fig. 1 are the current density–voltage (J – V) characteristics, current efficiency–current density characteristics, and operational stabilities of Devices A1, B1, C1, and D1. These devices have the same layer structure of ITO/75 nm NPB/30 nm Alq₃:C545T/30 nm Alq₃/Mg:Ag. However, each device has a different ITO surface condition: Device A1 has an as-prepared ITO, which was also stored for more than 30 days in an ambient (denoted as “as-prepared”); Device B1 has a washed ITO: the as-prepared ITO was washed by a commercial glass scrubber tool followed by oven baking and an optical inspection (denoted as “washed”); Device C1 has an O₂-treated ITO: the washed ITO was further treated by oxygen plasma (denoted as “O₂-treated”); and Device D1 has a CF_x-treated ITO: The O₂-treated ITO was further followed by a CF_x plasma treatment resulting in a ~1-nm-thick nonconductive CF_x layer onto the ITO surface (denoted as “CF_x-treated”).

As is expected, different ITO surface conditions result in quite different EL performances in the OLEDs. Shown in Fig. 1a, Device A1, having the as-prepared ITO, needs higher than 9 V when operated at 20 mA/cm². Compared to Device A1, Device B1, having a washed ITO, can reduce the drive voltage by more than 1 V at 20 mA/cm². After oxygen plasma treatment of the ITO surface, Device C1 further reduces the drive voltage to 6.9 V at 20 mA/cm². Furthermore, Device D1 with both oxygen and CF_x plasma treated ITO exhibits the lowest drive voltage. The voltage difference between Devices A1 and D1 is 3 V at 20 mA/cm². It is noticed that the cleanness of the

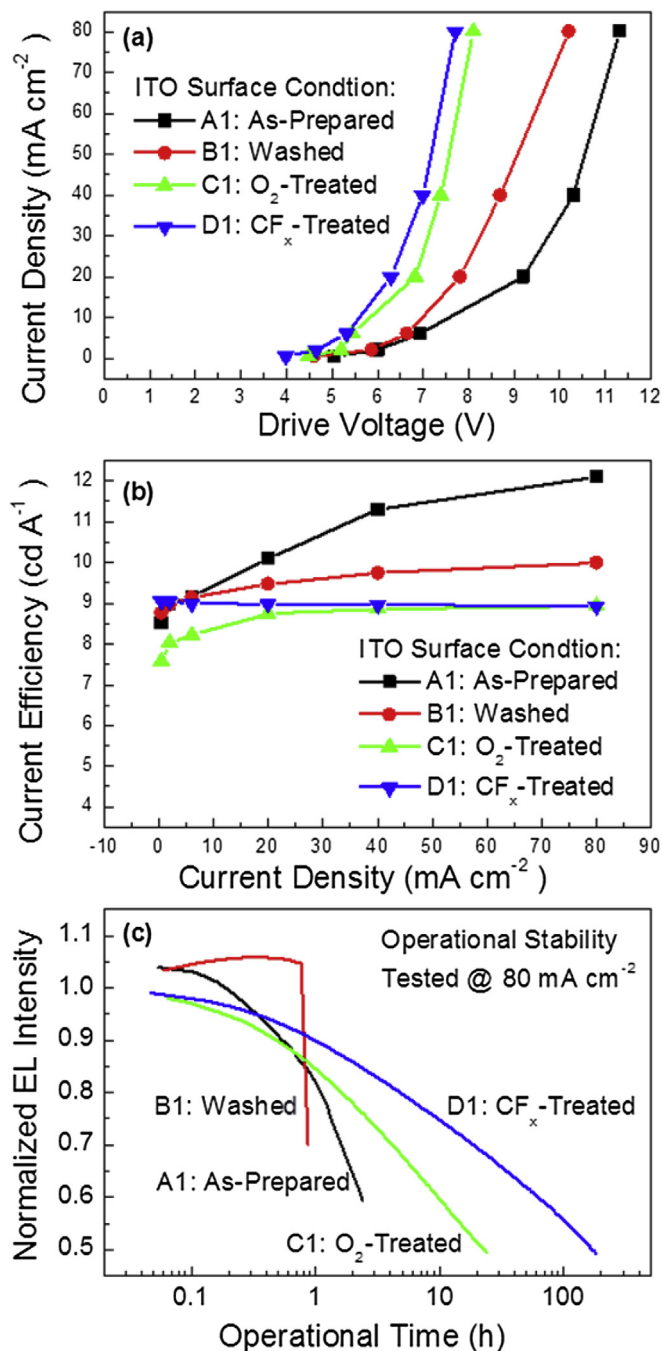


Fig. 1. a) Current density–voltage characteristics, b) current efficiency–current density characteristics, and c) operational stabilities of Devices A1, B1, C1, and D1. The layer structure of the devices is ITO/75 nm NPB/30 nm Alq₃:C545T/30 nm Alq₃/Mg:Ag. The ITO surface conditions of Devices A1, B1, C1, and D1 are as-prepared, washed, O₂-treated, and CF_x-treated, respectively.

ITO surface is gradually enhanced from Devices A1 to D1, which may result in the increase in the work function of the ITO and consequently the reduction in the hole injection barriers of the devices. Therefore, the drive voltage of OLEDs decreases from Devices A1 to D1. The difference in current efficiency between the devices is also apparent, as shown in Fig. 1b. Compared to Devices C1 and D1, Devices A1 and B1 have higher current efficiency. The higher current efficiency is due to the higher hole-injecting barrier at the interface between the ITO anode and the NPB layer (i.e., reduced hole current) in Devices A1 and B1. Because holes are

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