Materials and Design 160 (2018) 572–577

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Ag nanowire electrode with patterned dry film photoresist insulator for flexible organic light-emitting diode with various designs

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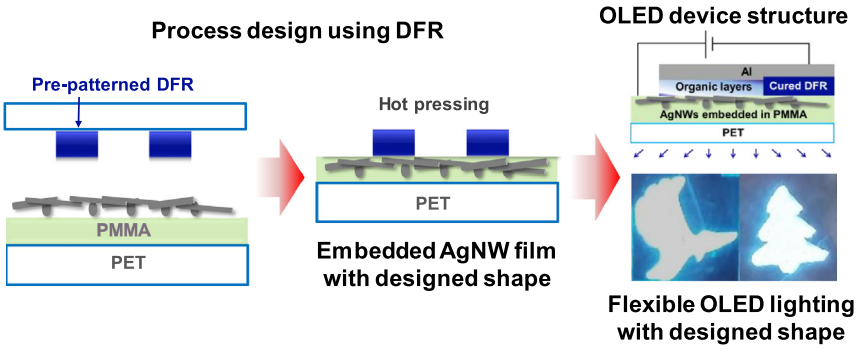
HIGHLIGHTS

- Ag nanowire flexible transparent electrodes with root mean square below 5 nm were fabricated by a single hot pressing process
- By using pre-patterned dry film photoresist, shaped Ag nanowire electrodes were fabricated in the same hot pressing process
- Flexible organic light-emitting diode lightings with various designs were realized with the fabricated electrodes
- Paper-based organic light-emitting diode lighting was realized using a dry film photoresist planarization layer

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 April 2018
Received in revised form 27 September 2018
Accepted 28 September 2018
Available online 02 October 2018

Keywords:
Dry film photoresist
Silver nanowire
Insulator
Electrode
Paper
Organic light-emitting diode

ABSTRACT

In this study, the utilization of a dry film photoresist (DFR) was explored for fabricating a patterned insulating layer in Ag nanowire (AgNW) bottom electrodes with various designs for flexible organic light-emitting diodes (OLEDs). With a single hot-pressing step, a pre-patterned DFR insulator was attached onto the AgNW electrodes, thus forming insulated electrodes in desired patterns. In the same hot-pressing step, AgNWs were also embedded into poly(methyl methacrylate), which sufficiently reduced the root mean square (RMS) roughness (~5 nm) for use in OLED lighting. Using DFR, AgNW electrodes with various shapes were fabricated to realize shaped flexible OLED lighting. The performance of OLED lighting employing the AgNW/patterned-DFR insulator electrode was comparable to that of indium-tin-oxide-based OLED lighting. Further, the DFR insulator was utilized as a planarization layer in a paper-based OLED lighting, which operated well with a turn-on voltage of 3.5 V and maximum luminance of 157 cd/m^2 at 7.5 V.

1. Introduction

With increasing interest in flexible organic light-emitting diode (OLED) lightings, the development of flexible transparent electrodes has become crucial for securing the reliability of devices under harsh mechanical strains [1–6]. Indium tin oxide (ITO) is commonly used for the electrodes of OLEDs; however, its intrinsic brittleness and high processing temperature limit its use in flexible OLEDs built on polymer substrates [7,8]. As alternatives of ITO, various material systems such as metal grids [9,10], carbon nanotubes [11,12], graphene [13,14], and metal nanowires [15–20] have been reported. Among these, Ag nanowires (AgNWs) are the most promising alternative owing to their high conductivity and transparency, which are comparable to those of ITO

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https://doi.org/10.1016/j.matdes.2018.09.051
0264-1275/

[21,22]. In addition, AgNWs have attracted the attention of electronics industry owing to their potential for scalable synthesis and ease of formation of large-scale conductive films on polymer substrates via simple coating methods such as bar coating, slot-die coating, and spray coating [23–26].

However, for OLED applications, the roughness of AgNW networks should be lower than ~10 nm. Stacked one-dimensional AgNWs on a substrate typically have a high surface roughness of more than a few hundred nanometers, where concentrated current causes the degradation of OLED performance; consequently, a dark spot appears on the OLED lighting panels. Typically, a reduction in the roughness of AgNW electrodes is achieved by embedding AgNWs into the surface of polymeric resins [27]. In the embedding process, AgNWs are first coated on a substrate, and then, a UV or thermally curable resin is deposited on the AgNWs and cured. During curing, AgNWs are bound to the surface of the resins. By detaching the cured resin from the substrate, a AgNW electrode embedded in the resin is fabricated. In addition, the embedding process can remarkably reduce the roughness of the AgNW to ~5 nm [27,28], which is sufficient for OLED lighting. However, the use of embedding process in the industry is limited owing to its complexity, as it includes curing and detachment steps, and the potential loss of conductivity during the detachment process. Moreover, an additional step to coat the insulating layer is required to obtain shaped or designed OLED lighting panels [29]. The typical liquid-type resin used for the insulating layer not only increases the complexity of the fabrication process with additional coating and curing steps, but also hinders its use in electrodes on a paper-based substrate. The increased number of fabrication steps as well as the limited application area due to the complexity of the aforementioned embedding process and the liquid resin-based insulating process increases the processing and material costs, thereby limiting the use of AgNWs in the flexible OLED lighting industry. Therefore, a new process design for the simple fabrication of flexible and transparent AgNW electrodes with low roughness below ~10 nm and good compatibility with various substrates, including paper-based substrates, should be developed to widen the application areas of AgNW electrodes in the flexible OLED lighting industry.

In the present study, a simple and cost-effective fabrication process to prepare AgNW electrodes with low roughness for flexible and patterned OLED lighting is designed using a simple hot pressing technique with a dry film photoresist (DFR) insulating layer. DFR is a solid-film-type negative photoresist that can be attached on any type of substrate, such as polymer, glass, paper, and ceramic, by simple lamination or hot pressing. Herein, AgNW electrodes with designed patterns were fabricated by using a pre-patterned DFR, and the roughness of the AgNWs was remarkably reduced by embedding the AgNWs into poly(methyl methacrylate) (PMMA) having a low melting temperature (T_m). AgNW electrodes embedded in PMMA with the designed patterns of an insulating layer were easily fabricated using a single hot-pressing step without requiring complex coating and detachment processes or any additional step to form the insulating layer. The shaped OLED lighting employing the AgNW/patterned-DFR insulator electrode demonstrates good performance, comparable to that of ITO-based OLED lighting. Furthermore, the utilization of DFR insulator for OLED lighting has also been demonstrated for paper-based OLED lighting.

2. Experiment

2.1. Materials

PMMA powder (Aldrich, M.W.: 996,000) was dissolved in water with anisole ($CH_3OC_6H_5$, Daejung) to prepare a 4 wt% PMMA solution. AgNW suspension in isopropyl alcohol with 1 wt% solid content was purchased from Nanopixis Co. Ltd., and the average diameter and length of the AgNWs were ~35 nm and ~25 μm, respectively. Roll-type DFR films (KL-1015) with 50-mm length, 15-mm thickness, and 30-mm width were purchased from KOLON Industries. Hot press equipment was used to embed the AgNWs into PMMA.

To fabricate the AgNWs/PMMA/PET electrodes, a Mayer rod (#6, RD Specialist Inc.) was used to coat PMMA on the poly(ethylene terephthalate) (PET) substrate, and then, AgNWs were coated on the PMMA/PET substrate. Meanwhile, the DFR/cover film was UV-cured with a patterned photomask. Afterward, a Na_2CO_3 aqueous solution (1 wt%) was used to develop the un-cured region of DFR. The DFR/cover film and the AgNWs/PMMA/PET film were then hot-pressed at 150 °C under 40 MPa for 5 min.

For the paper-based electrode, a silver nanoparticle (AgNP) suspension in ethanol with 50 wt% solid content was purchased from N&B, and the average diameter of the AgNPs was ~50 nm. A super-hydrophobic PET (SHP-PET) film (SKC, thickness: 25 μm), paper (milk, A4 80 g), black toner (HP, CF210A), and office laser printer (HP, M276nw) were used in the dry transfer process.

To prepare the paper-based electrodes, the dry transfer method using toner-printed paper was used. First, AgNPs were coated on the SHP-PET film. Then, the AgNPs on SHP-PET were placed on the toner-printed paper and hot-pressed to transfer and embed them in the toner. The DFR prepared using the same method as that used to prepare the AgNPs/PMMA/PET film was subsequently placed and hot-pressed on the AgNPs/toner/paper electrodes, thereby forming a patterned insulating layer. The detailed mechanism of the transfer process using the toner-printed paper is reported elsewhere [30].

2.2. OLED fabrication

To fabricate a bottom-emitting blue OLED, the following layers were thermally evaporated on the DFR/AgNWs/PMMA/PET film and ITO glass: 1,4,5,8,9,11-Hexaazatriphenylene-hexacarbonitrile, 60 nm/DS-HTL, 40 nm/2-methyl-9,10-di(2-naphthyl) anthracene: 5 wt% 1,6-bis (*N*-phenyl-p-CN-phenylamino)-pyrenes, 20 nm/LG-201:Liq (1:1), 30 nm/Liq, 1.5 nm/Al, 100 nm. To fabricate a paper-based top-emitting blue OLED, 10-nm-thick Al layer and 10-nm-thick Ag layer were deposited using a shadow mask; these layers functioned as semi-transparent top electrodes. Organic layers and a cathode metal layer were deposited under 8×10^{-7} Torr in a vacuum evaporator with deposition rates of 0.1 and 1 Å/s, respectively. The current–

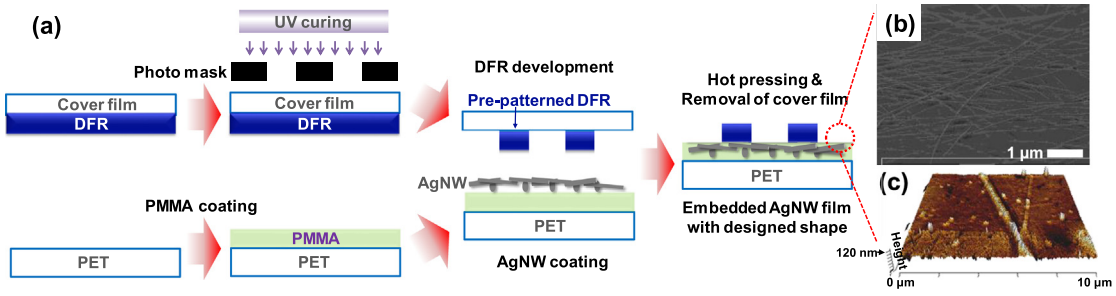

Fig. 1. (a) Schematic illustrations of fabrication step of patterned and embedded AgNW electrodes with DFR insulating layer. (b) 35° tilted SEM image and (c) AFM image of AgNWs embedded into PMMA.