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Organic wrinkles for energy efficient organic light emitting diodes

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ABSTRACT

Extracting the confined light is of critical significance in achieving highly energy efficient organic light emitting diodes. To address the task of extracting the confined light, we here synthesize a new type of liquid prepolymer, which spontaneously forms wrinkles upon ultra-violet light exposure. The spontaneously formed organic wrinkle is successfully applied not only in extracting the confined light but also in inducing angular spectral stability. Simulations demonstrate that the wrinkles can lower incident angle of light impinging on the substrate/air interface and thus help extract a large portion of light delivered to the substrate. In particular, it is shown that geometrical optimization of the size and aspect ratio of wrinkles is important in obtaining the highest light extraction. With the simplicity of the process and size controllability, the proposed wrinkle-based approach can be readily realized over a large area, opening up a new avenue in various photonics applications.

1. Introduction

The efficiencies of light emitting and harvesting devices may be enhanced by optical manipulations. Lighting energy policy is directed toward improving efficiency to lower the energy consumption and emission of greenhouse gases. Currently, approximately 20% of the global electricity is used for residential and commercial lighting [1]. In this context, enhancing the efficiencies of lighting apparatus has emerged as a crucial task. Organic light emitting diodes (OLEDs) are regarded, along with other solid-state light-emitting devices, as a light source that can respond to the compelling needs for energy efficient and environmentally sound light sources for their human friendly self-emissive character, relatively low energy required for fabrications and many more merits [2–8]. However, due to the presence of optical obstacles in OLEDs, only a limited fraction (≈20%) of the generated light can be out-coupled, while the rest is wave guided, confined in a substrate, or lost to parasitic absorption of surface plasmon polariton modes [9]. The optical method related to the light retrieving and enhancing efficiency is referred as light extraction techniques [10]. Because the light extraction can lead to improved efficiency and lower power consumption, light extraction emerges as a crucial technology in energy efficient OLED applications. Various OLED light extraction methods have thus been proposed. Technical strategies commonly involve the introduction of microstructures which can contributed in deflecting the light traveling path or increase the internal scattering. Broadly, OLED light extraction can be divided as external and internal methods. In the external method, the external surface of the substrate is modified for extraction of substrate modes or the light confined within substrates [11–15]. The internal method focuses on the extraction of the waveguided modes or the light guided along the electrode (e.g. ITO)/organics layers [16–19]. In order to achieve high light out coupling effects various approaches have been suggested. To extract the waveguided light in top emission OLED polymeric microlens were molded on the top emitting surface [20]. A corrugated sapphire substrate has been applied to harvest the lost associated to the surface plasmon polariton [21]. Light extracting layer based on polymer-metal oxide composite was applied to make OLEDs energy efficient [22]. Phase separation of binary polymer blend was utilized to fabricate light out coupling structures [23]. These methods have a common feature of bringing plural light extraction methods into one entity. Although significant advancements have been achieved in forming light extraction structures, the necessities of vacuum deposition, complex patterning methods and precise size/spacing negate many light extraction strategies, especially over large areas.

In this work we synthesized a new liquid prepolymer which spontaneously forms spontaneously wrinkles upon a single step light exposure at room temperature. Our approach obviates the

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Table 1
The full chemical names and their functions of organics used in OLED.

Abbreviation	Full chemical name	Function
Hat-CN	1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile	Charge generation layer (*n*-type organic semiconductor)
TAPC	1,1-bis[(di-4-tolylamino)phenyl]cyclohexane	Hole transport layer
DCzPPy	2,6-bis(3-(carbazol-9-yl)phenyl)pyridine	Emitter host
Ir(ppy)III	Tris(2-phenylpyridine)Iridium	Emitter dopant (phosphorescent green)
TmPyPB	1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene	Electron transport layer

technical difficulties involved in fabricating conventional light extraction structures and can be readily applied to large areas. We applied wrinkle as an external light extraction structure for achieving energy efficient OLEDs. We investigated the wrinkle size effect on the light out coupling and performed optical simulations to elucidate the morphological factors which are important in achieving the highest light extraction.

2. Experimental section

The formation of wavy wrinkle patterns is a geometric effort of a material system to stay in its lowest energy configuration [24,25]. Wrinkles are produced when difference in in-plane compressive stress is present across the film thickness. If the in-plane stress in a geometrically constrained film exceeds a critical value wrinkles forms spontaneously [24]. In forming wrinkles, we use a liquid prepolymer as a starting material. This is in contrast to a frequently employed method, in which two materials of different mechanical properties are stacked. It is noteworthy that a material in liquid state, rather than not solid state, is used; this can be advantageous in terms of easy (and thus low-cost) processing. Our liquid prepolymer is linear and has two moieties at each the molecular terminal. The chemical structure can be found in Supporting Information [S1]. The proposed wrinkle forming process consisted of two-steps. In the first step, liquid prepolymer was coated to form a liquid thin film. In the second step, organic thin films were exposed to a UV light, during which wrinkles form spontaneously. In order to facilitate the UV process a small amount (1.5 wt.%) of commercial photoinitiator (Irgacure 184, Ciba) was added to the liquid prepolymer. Due to the consumption of photo initiator radicals by oxygen, UV exposure in an atmospheric environment did not yield wrinkles. Thus, it is essential to carry out the exposure step in a N_2 atmosphere. The transmittances and hazes of wrinkle films were measured using a UV–visible spectrophotometer (U-3501, Hitachi) a haze meter (Haze-gard plus, BYK), respectively. The morphologies of the wrinkles were investigated by means of scanning electron microscopy (SEM, Sirion 400 FEI) and atomic force microscopy (AFM, XE-100, Park System).

In order to evaluate our wrinkles as a light extraction functional, we have fabricated bottom-emission type phosphorescent green OLEDs, which have an emission area of $10 \times 7\ mm^2$. The anode and the cathode were indium tin oxide (ITO) and LiF/Al, respectively. In this work we have thermally deposited organics to fabricate OLEDs. Our phosphorescent green OLEDs have a stack structure of ITO (70 nm)/Hat-CN (5 nm)/TAPC (30 nm)/Hat-CN (5 nm)/TAPC (30 nm)/Hat-CN (5 nm)/TAPC (30 nm)/DCzPPy: Ir(ppy)III (7%) (20 nm)/TmPyPB(30 nm)/LiF (1 nm)/Al(100 nm). For the hole transport layer (HTL), we have adopted a layered HTL of Hat-CN and TAPC. Such layered structure has been shown not only to enhance the hole transport but also electrically stabilize the device [27]. The full chemical names and their functions are listed in Table 1. The base pressure of all deposition processes were below 6.66×10^{-5} Pa. In order to prevent accidental device degradation all OLEDs were glass encapsulated in a N_2-filled glove box. The electroluminescence (EL) spectra were collected using a spectroradiometer (CS-2000, Minolta) equipped on a goniometer. The current density–voltage (*J*–*V*) and luminescence–voltage (*L*–*V*) characteristics were measured with a current/voltage source/measure unit (Keithley 238) and a spectroradiometer (CS-100, Minolta), respectively. Wrinkles films of different sizes were prepared on polyethylene (PET) films, which have glass index matching adhesive on one side. The films were obtained from Kureha. The PET films with the wrinkles were then attached to the emission surface with the wrinkle side facing air.

3. Results and discussion

Fig. 1 shows scanning electron microscopy (SEM) images of wrinkles as a function of spin-coating speed or film thickness. Recalling the wrinkle formation mechanism, the formability of wrinkles by a UV process in a single liquid prepolymer system strongly indicates the formation of a hard skin on a soft foundation.

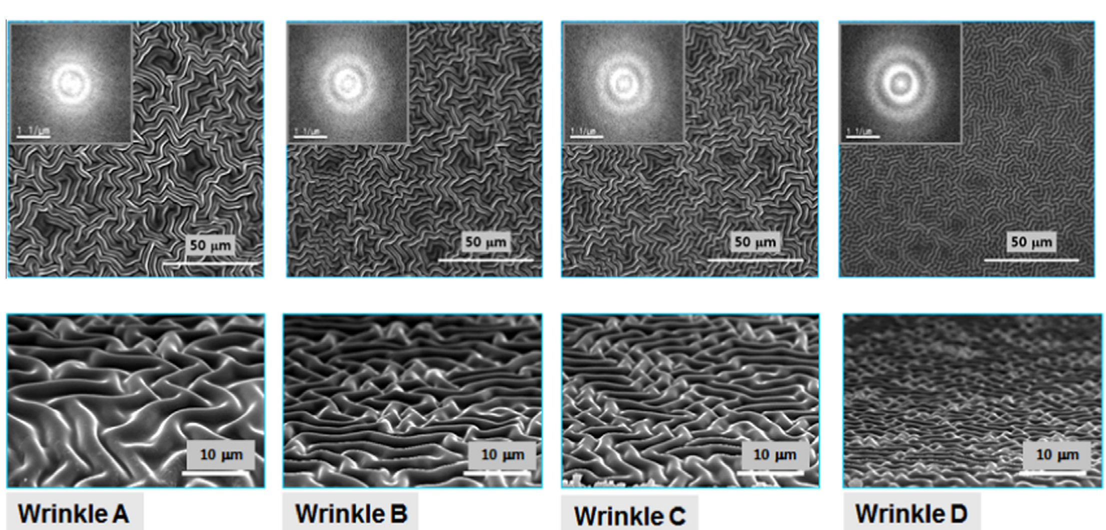

Fig. 1. SEM images of wrinkles as a functions of spin-coating speed. Insets are the FT images of the corresponding SEM images.