



Surface planarization effect of siloxane derivatives in organic semiconductor layers



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ABSTRACT

The ability of siloxane surface control additives (SCAs) to planarize organic semiconductor films with a thickness of tens of nanometers printed on indium tin oxide (ITO) surfaces with stripe-patterned bank structures using a liquid-phase method is demonstrated. Three types of SCAs with different molecular structures are examined in organic solutions of toluene, anisole and tetralin containing N,N'-bis(3-methylphenyl)-N,N'-bis(phenyl)-benzidine as a solute and typical organic semiconductor. While there is an optimum SCA and concentration for each solution, one type of SCA is comprehensively effective for all solutions. This SCA increased contact angle, which is contrary to the typical behavior of SCAs. Scanning electron microscope images of the thin films near the banks reveal that this SCA did not change the contact area between the film and substrate surface, which is related to the effectiveness of the SCA. SCAs did not affect the current–voltage characteristics of green organic light-emitting diodes, but did increase external quantum efficiencies, suggesting that SCAs can be used to improve the quality of solution-deposited films for use in optical devices.

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1. Introduction

Solution processes to fabricate organic devices such as organic light-emitting diodes (OLEDs), organic transistors, and organic solar cells have attracted much attention for many years [1–3] because of their wide variety of applications. In particular, to produce large OLED televisions, development of independent RGB fine patterning is needed to realize low-cost mass production because of process capabilities on large glass substrates (Gen 8 or larger). Many solution processes, including inkjet [4], nozzle jet [5], and gravure [6] printing, have been developed during the last decade for large-scale applications. However, numerous issues still limit the commercial use of these solution processes. One of the most important issues is to ensure inter- and intra-pixel uniformity, which is not achieved when the solvent vaporization of inks is nonuniform.

The behavior of a drying droplet is well known as the coffee-ring effect [7], the mechanism of which has been widely studied using polymer particles and nanoparticle dispersion systems [8,9]. The formation of coffee rings is caused by the outward flow during solvent evaporation after the contact line is pinned. Particle dispersion systems are often used to overcome coffee-ring formation; for instance, solvent mixing [10–12], modification of solute shape [13], and

addition of surfactants. Various surfactants have been used to achieve uniform morphology. Kajiya et al. [14] investigated the influence of a fluorosurfactant on film morphology using monodisperse polystyrene dissolved in dipropylene glycol methyl ether acetate with a bank structure. They reported the accumulation of surfactant near the solution edge generated reverse Marangoni flow, which resulted in flat polymer films with a thickness of around 2 μm. Other surfactants such as sodium dodecyl sulfate [15] and polyether-modified polydimethylsiloxane have also been investigated in water-based polystyrene dispersion systems [16]. However, there are few reports examining the film morphologies of organic solution-processed semiconductor devices. Organic devices require films with thicknesses of around tens of nanometers and uniform film thickness (variation within several percent) to achieve acceptable performance. The organic materials are inherently insulating, so controlled film thickness is required for effective carrier transport and to avoid breakdown caused by current concentration in thinner areas.

In this article, we focus on the morphology of thin films (tens of nanometers thick) of small-molecule organic semiconductor materials that are homogeneously dissolved in single organic solvents with surface control additives (SCAs). The effect of SCAs on striped films printed on indium tin oxide (ITO) surfaces between bank structures using a dispenser is investigated. We chose aralkyl- and polyether-modified methyl siloxane derivatives as SCAs because they are insulating with wide bandgaps, so they are not expected to markedly affect OLED performance.

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2. Experimental

2.1. Substrates

Stripe-patterned bank structures were prepared by conventional photolithography. A negative-working photosensitive material based on polyimide (ATX5901Pb-1, Asahi Glass Co., Japan) was spin coated on a cleaned ITO-coated glass substrate. The substrate was baked for 2 min at 100 °C, photodecomposed under irradiation with hybrid light with wavelengths of 365, 405 and 436 nm through a photomask with lines for 20 s, soaked in a developer (2.38 wt.% tetramethylammonium hydroxide in methanol) for 1 min, and finally washed with pure water. The cross sectional view of the substrate was demonstrated by a scanning electron microscope (SEM: SU8000, Hitachi High Technologies Co. Japan) shown in Fig. 1.

To measure contact angle, a flat surface of the bank material was prepared following the same procedure, except that the substrate was irradiated without the photomask. Both substrates were cleaned with ultraviolet/ozone cleaner (UV253, Filgen Inc., Japan) for 12 min immediately before measurement.

2.2. Coating liquids

N,N'-Bis(3-methylphenyl)-N,N'-bis(phenyl)-benzidine (TPD; Tokyo Chemical Industry Co., Ltd., Japan, 97% purity) was chosen as a representative solute because of its good solubility in various solvents. Another reason for its selection is because TPD films have amorphous structures [17] which should give a smoother surface than a film with crystalline structure.

Three different solvents, toluene (Wako Pure Chemical Industries, Ltd., Japan, 99.5%, boiling point bp = 111 °C, surface tension γ = 29.7 mN/m, viscosity η = 0.590 mPa·s), anisole (Wako Pure Chemical Industries, Ltd., 99%, bp = 154 °C, γ = 34.6 mN/m, η = 0.984 mPa·s), and tetralin (Kanto Chemical Co. Inc., Japan, 98%, bp = 207 °C, γ = 35.3 mN/m, η = 2.14 mPa·s), were used.

The chemical structures of the siloxane SCAs used here are presented in Fig. 2. BYK322 is a polymeric, aralkyl-modified alkyl methyl siloxane with a molecular weight (Mw) of ca. 10,000, BYK307 is a polymeric,

polyether-modified dimethyl siloxane (Mw ca. 10,000), and BYK348 is a monomeric, polyether-modified dimethyl siloxane (Mw ca. 1000). The number of ethylene oxide units in each polyether chain of BYK307 is 40–50, and the units are attached at both ends of the molecule. BYK348 has one long-chain polyether unit (10–20 ethylene oxide units) attached at the center to a silicon atom. All SCAs, viscous liquids with purity >97%, were provided by BYK Japan KK and used without further purification.

Coating liquids were prepared by dissolving TPD in one of the solvents described above, and subsequently adding a SCA. TPD mass concentrations were 1 and 2 wt.%. The mass concentrations of the SCAs were in the range of 0.005–0.05 wt.%. The addition of up to 5 wt.% TPD had little effect on the viscosity and surface tension of the solvent.

2.3. Printing method

A dispenser system (Shotmaster SM500DS, Musashi Engineering Co., Japan) was used as a printing tool because of its wide process capability compared with that of an inkjet printer. This system was composed of an air compressor for jetting, CCD camera to aid positioning, a nozzle mounting unit, and a PC controller. The diameter of the nozzle was 10 μ m. The inks were continuously injected on the striped bank substrate placed on a stage horizontally moving at 900 mm/s. The printing was made every five columns. The flow rate of the injected liquid was controlled by the pressure applied to an ink tank from the air compressor.

After drying the film at 25 °C for 1 h, the surface profile including the ITO surfaces on the both side of coated surface was measured using an optical surface profiler (NewView 7300, Zygo, USA). The profile of the film was corrected referencing the vertical levels of the ITO surfaces.

Considering 2 wt.%-TPD solution coating on the ITO surface with 90 μ m width, 0.002 g/s of the flow rate was selected to fabricate a film with the thickness of 50 nm. However, the flow rate was very sensitive to the pressure in this range because it was near a low limit of the nozzle ability. As a result, the thicknesses varied between 40 and 100 nm.

2.4. OLED device

The OLED devices had the structure ITO (100 nm)/hole injection layer (HIL; 30 nm)/interlayer (IL; 20 nm)/emitting layer (EML; 50 nm)/electron transporting layer (ETL; 30 nm)/electron injection layer (EIL; 0.8 nm)/cathode (100 nm). The materials used for each layer were ELsource SHI-2520-IJ8 (Nissan Chemical Co., Japan) as the HIL, thermally cross-linked polyfluorene derivative HTI (Sumitomo Chemical Co., Japan) as the IL, and small-molecule host material 1,3-bis(N-carbazolyl)benzene (mCP; Luminescence Technology Co., Taiwan, R.O.C., >99.5% purity) containing 5 wt.% of green fluorescent guest 9,10-bis[N,N-di-(p-tolyl)- amino]anthracene (TPPA; Luminescence Technology Co., >99% purity) [18] as the EML. Here, mCP was employed as a host material because of its wide bandgap and common use as a host material [19]. Because the chemical structures of mCP and TPD are similar in terms of Mw and chemical moieties, these materials were expected to exhibit similar physical behavior when a SCA was added to the EML layer. HIL, IL and EML were spin coated and annealed under nitrogen atmosphere followed by vacuum deposition of tris (8-hydroxyquinolino) aluminum (Alq₃), LiF and Al as ETL, EIL and cathode, respectively. Each SCA was added to the EML ink to examine the influence of each SCA on OLED characteristics. A source measure unit (B2912A, Agilent Technologies, USA) and spectroradiometer (CS2000A, Konica Minolta Inc., USA) were used to obtain initial current–voltage characteristics and external quantum efficiency (EQE) measurements assuming a Lambertian distribution.

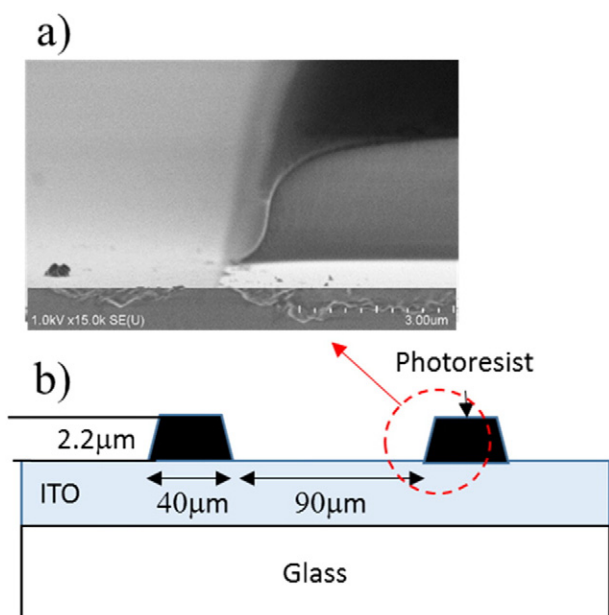


Fig. 1. (a) SEM image of a polyimide bank. (b) Cross-sectional illustration of a substrate with banks.

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