



The optimisation of the laser-induced forward transfer process for fabrication of polyfluorene-based organic light-emitting diode pixels

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ABSTRACT

Laser-induced forward transfer (LIFT) has already been used to fabricate various types of organic light-emitting diodes (OLEDs), and the process itself has been optimised and refined considerably since OLED pixels were first demonstrated. In particular, a dynamic release layer (DRL) of triazene polymer has been used, the environmental pressure has been reduced down to a medium vacuum, and the donor receiver gap has been controlled with the use of spacers. Insight into the LIFT process's effect upon OLED pixel performance is presented here, obtained through optimisation of three-colour polyfluorene-based OLEDs. A marked dependence of the pixel morphology quality on the cathode metal is observed, and the laser transfer fluence dependence is also analysed. The pixel device performances are compared to conventionally fabricated devices, and cathode effects have been looked at in detail. The silver cathode pixels show more heterogeneous pixel morphologies, and a correspondingly poorer efficiency characteristics. The aluminium cathode pixels have greater green electroluminescent emission than both the silver cathode pixels and the conventionally fabricated aluminium devices, and the green emission has a fluence dependence for silver cathode pixels.

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1. Introduction

The use of laser-induced forward transfer (LIFT) to deposit functional materials is a field growing rapidly [1]. From the earliest research into laser-deposited inks [2] and metals [3], the variety and complexity of the materials investigated has increased exponentially. The first use of a sacrificial intermediate 'dynamic release layer' (DRL) to help with the laser ablation process was in 1993 [4], but this used a thermally-ablated black carbon layer. Thermal DRLs were the original focus, and a technique evolved called laser-induced thermal imaging (LITI), with a particular application for organic thin-film layers in OLEDs [5] and other organic electronics. The use of a photochemically sensitive polymer layer to reduce the thermal load on the transfer layer was first applied with the same triazene polymer (TP) used in this study, to transfer living

neuroblast cells [6]. Since then, TP-based LIFT has been applied to quantum dots [7], polymer LEDs (PLEDs) [8–10], biological solutions [11], ceramics [12], polymeric sensors [13], organic thin-film transistors [14], and polystyrene microbead arrays [15].

Previously, functional OLED pixels have been printed side-by-side with a controlled gap and at reduced environmental pressure [10], using the results from reduced pressure and substrate-substrate gap optimisation of the LIFT process [16]. The results presented here are a bridge between these two studies, demonstrating some material and laser effects upon pixel morphologies and performance. The TP layer thickness was investigated before, and it was concluded that a thickness of at least 150 nm was required [16]. In combination with suitably adhesive receiver substrates [9], this has lowered the required fluence for successful transfer of pixels from over 200 mJ/cm² to well under 100 mJ/cm². All the pixels presented here were transferred between 50 and 105 mJ/cm², meaning that the pixel morphological effects observed in this research have occurred at low fluences relative to many other LIFT studies, even with a TP DRL (above 200 mJ/cm², e.g. Refs. [13,14]).

Ag was used as the metal layer in the transfer optimisation studies [16], both because it is an easier metal to evaporate than Al and because it gives better device performances of conventionally fabricated devices (see Table 1). Hence in this study, Ag was chosen

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Table 1
Device performances for various conventionally fabricated devices, at a current density (CD) of $\sim 40\text{--}50\text{ mA/cm}^2$. Blue, green and red refer to the LEP layer, whether just plain PFO (blue), or doped PFO (red and green).

Colour	Cathode	Bias [V]	CD [mA/cm^2]	Lum [cd/m^2]	LE [cd/A]
Blue	Ag	19	42.7	29.4	0.069
	Ag/ Cs_2CO_3	14	53.2	816.9	1.54
	Al	21	31.1	0.73	0.0023
Green	Ag	32	43.1	391.2	0.91
	Ag/ Cs_2CO_3	22	36.9	885.2	2.4
	Al	37	48.3	134.6	0.28
Red	Ag	33	48.6	114.1	0.24
	Ag/ Cs_2CO_3	21	30.8	299.1	0.97
	Al	42	41.7	49.8	0.12

as a cathode material, and Cs_2CO_3 was also added to the Ag to try and create devices with better electron injection [17]. Because Al had been often used before, and observed to give better LIFT pixel device characteristics than conventionally fabricated devices in some cases [8,9], it was also chosen. Fig. 1 shows the optimisation progression, starting with (a) the first simple polyfluorene (PFO) pixels transferred at the optimised conditions from Ref. [16], then the first R–G–B pixels with Ag cathodes (b), and Al cathodes (c), then finally the optimised pixels from Ref. [10], with Al cathodes (d), and Al/TBA (tetrabutyl ammonium hydroxide) (e). The present article focusses on the pixels from Fig. 1b and c in particular.

2. Experimental

The donor substrates are fused silica substrates ($25\text{ mm} \times 25\text{ mm} \times 1\text{ mm}$), spin-coated with a 190 nm triazine polymer (TP) film (synthesised in-house [18]), followed by an evaporated 80 nm cathode of either silver or aluminium, with the thickness measured using a calibrated quartz crystal microbalance. The Ag cathodes were further modified by the evaporation of a thin layer of caesium carbonate (Cs_2CO_3) onto the Ag, which is a commonly used electron-injecting layer [17]. On top of the cathode, the light-emitting polymer (LEP) layer was poly(9,9-dioctylfluorenyl-2,7-diyl) capped with silsesquioxane (PFO, bought from ADS), spin-coated from a toluene:*p*-xylene (1:1) solution, and doped

with red (2 wt%) and green (10 wt%) emitting iridium dyes (see Ref. [10] for details). The PFO films of the devices presented here were all 80 nm ($\pm 10\text{ nm}$) thick, except for the optimised Al pixels, from the previously published three-colour OLED study [10], which had 50 nm thick PFO films.

The receiver substrates are patterned ITO on glass ($25.4\text{ mm} \times 25.4\text{ mm} \times 1\text{ mm}$), spin-coated with 60 nm PEDOT:PSS (CleviosTM P Al 4083), then 40 nm PVK ($M_w = 1,100,000$, Sigma–Aldrich). For conventional devices, the PFO was spin-coated onto the PVK followed by the evaporation of the cathode material (Al, Ag or $\text{Cs}_2\text{CO}_3/\text{Ag}$). All glass substrates were cleaned thoroughly prior to film deposition, according to the procedure outlined before [16], and the samples were kept in an inert nitrogen environment throughout film deposition. All spin-coated film thicknesses were measured using a profilometer (Ambios XP-1).

The LIFT experiments were carried out with a steel spacer between the donor and receiver, giving a $\sim 15\text{ }\mu\text{m}$ gap measured by interference in UV–vis–IR spectroscopy [16]. The samples were placed in a vacuum chamber, and the pressure reduced to 1 mbar. Single pulses from a XeCl excimer laser ($\tau \approx 30\text{ ns}$, $\lambda = 308\text{ nm}$) were obtained using a mechanical shutter, a rectangular aperture and an imaging lens to create a $\sim 0.5\text{ mm} \times 1.2\text{ mm}$ ablation crater in the TP layer. The laser fluence was controlled using a variable attenuator plate, and measured with a pyroelectric energy detector (Gentec QE12).

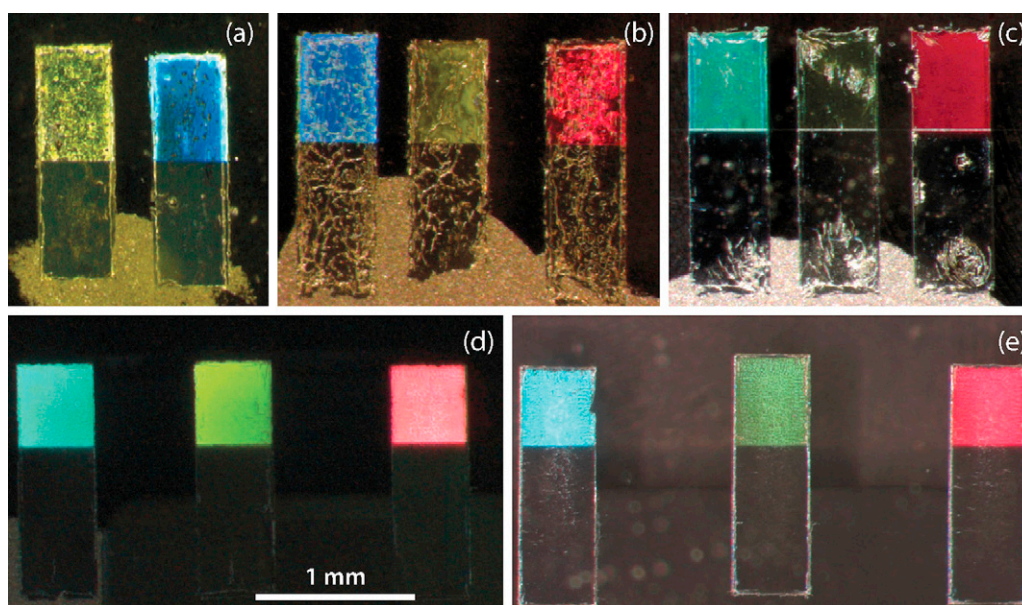


Fig. 1. Progress in the transfer of multiple PFO PLED pixels by LIFT. The first transfers of plain blue and green transfers with Ag/ Cs_2CO_3 cathode are shown (a), then the first three colour pixels with a Ag cathode (b), then the preliminary pixels with an Al cathode (c), before the optimised pixels with an Al cathode (d) and an Al/TBA cathode (e). All the pixels were transferred at approximately the same fluence, a fluence of $\sim 70\text{ mJ/cm}^2$.

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