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**PAPER**

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## Fabrication of solution-processable OFET memory using a nano-floating gate based on a phthalocyanine-cored star-shaped polymer†

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Solution-processed organic field-effect transistor (OFET) memory devices are fabricated using a blend film of 6,13-bis(triisopropylsilylethynyl)pentacene and phthalocyanine-cored star-shaped polystyrene. A highly crystalline organic semiconductor thin film was obtained on the star-shaped polymer with charge-trapping sites *via* a one-pot spin-coating process through vertical phase separation, which is advantageous for OFET memory device applications. The resultant OFET device demonstrated a charge carrier mobility of  $0.10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and an on/off current ratio of  $10^6$ . Upon application of a gate bias, a substantial reversible threshold shift was observed, along with long charge-retention ability, thereby confirming the memory characteristics of the device.

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### Introduction

Nonvolatile organic field-effect transistor (OFET) memory devices have attracted much attention because of their easily integrated structure and nondestructive readout.<sup>1,2</sup> They are regarded as a promising technology to integrate switching capacity and memory into various organic electronic devices such as sensors, displays, and radio-frequency identification tags. Meeting the recent demand for flexible, wearable, and large-area electronic devices *via* an easy and low-cost production process will necessitate the development of solution-processable OFET memories.<sup>3–5</sup> However, OFET memory often requires complicated and multistep fabrication processes because it contains multilayers of thin films, including an organic semiconductor for charge transport and a memory layer for charge trapping as well as a tunneling layer for charge insertion/extraction and a blocking layer for charge retention.<sup>6–8</sup> In addition, the active layer of an organic semiconductor in OFET memory devices is normally prepared by vacuum deposition to achieve high charge carrier mobility. Therefore, the preparation of OFET

memory devices using simple solution processes is a key challenge.<sup>9–14</sup>

Solution-processable organic semiconductors themselves have been extensively investigated and incorporated into OFET devices. For instance, 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS-pentacene) is a typical small-molecule semiconductor with solubilizing side groups. The charge-carrier mobility in OFET devices is strongly influenced by the crystalline structure and alignment of the organic semiconductors.<sup>15</sup> To achieve high device performance using a solution process, researchers have studied various methods of preparing thin films of organic semiconductors. Polymer-matrix-assisted phase separation is one of the most promising methods to prepare OFETs with high charge carrier mobility.<sup>16–21</sup> The mixed polymer increases the wettability of the substrates and decreases the evaporation speed of the solvents to induce crystallization of the organic semiconductor. For instance, an OFET prepared using a spin-coated blend film of TIPS-pentacene and poly( $\alpha$ -methylstyrene) exhibited a charge carrier mobility as high as  $0.54 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .<sup>17</sup> The selection of an appropriate polymer insulator,<sup>22–24</sup> polymer molecular weight,<sup>17,25,26</sup> and processing solvent<sup>27–30</sup> is important for controlling the crystal growth of organic semiconductors in the polymer matrix. Further advances in the coating process, such as the development of the blade coating technique,<sup>25,31,32</sup> have led to additional enhancements in OFET device performance.

Here we envision that the method of using a polymer matrix to construct high-performance OFET devices *via* a solution process can also be applied to OFET memory devices when the matrix polymer is capable of charge storage. Recently, we prepared metallophthalocyanine (MPc)-cored star-shaped

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polystyrene (MSP) and used it as a charge-trapping material in a pentacene-based OFET memory device.<sup>33</sup> The star polymer with an MPC core is readily soluble in organic solvents, enabling the fabrication of polymer films with a smooth surface *via* simple spin-coating onto a Si substrate. The MSP films exhibited a unique three-dimensional morphology, where the MPC cores were distributed and isolated by the surrounding arm-polymer matrix. A bottom-gate top-contact OFET memory device was fabricated *via* vacuum deposition of pentacene onto the polymer film, followed by deposition of Au electrodes. When a negative gate voltage was applied to the device, hole charges that accumulated in pentacene migrated to the MPC core, whereas polystyrene arms restrained leakage of the trapped charges to achieve nonvolatile memory similar to that based on nano-floating-gate transistors. In addition, the density of the MPC core—specifically, charge-trapping sites, can be controlled by the length of the arm polymer, enabling control of the resultant memory device performance.<sup>34</sup>

As a continuation of our studies on the OFET memory devices, we herein attempt to develop a simple and easy method of preparing OFET memory devices using polystyrene-based nano-floating gates. We used our unique design of memory materials—star-shaped polystyrene with specific charge-trapping sites—to prepare vertically phase-separated organic semiconductor/memory layers *via* a one-pot solution process. More specifically, the MPC-cored star-shaped polystyrene promotes the crystallization of small molecular semiconductors of TIPS-pentacene *via* polymer-matrix-assisted phase-separation and simultaneously provides organic semiconductor/memory layers on the substrates. The morphology and structure of the blend films of the star polymer and TIPS-pentacene prepared by a simple spin-coating process from various organic solvents were investigated using X-ray diffraction (XRD), atomic force microscopy (AFM), polarized optical microscopy, UV-vis absorption spectroscopy, and X-ray photoelectron spectroscopy (XPS). We obtained highly crystalline and vertically phase-separated TIPS-pentacene on star polymer blend films from an *ortho*-dichlorobenzene (ODCB) solution, which is advantageous for OFET memory device applications. We finally demonstrated an OFET device with a bottom-gate top-contact configuration (Fig. 1a).

The resultant device showed a hole mobility of  $0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and a high on/off current ratio greater than  $10^6$ . In addition, a memory window of 19.7 V was probed under an applied gate voltage in the range  $\pm 50 \text{ V}$  and the device demonstrated long charge-retention ability. Thus, the present one-pot solution process would be a useful approach for preparing non-volatile organic memories without cumbersome fabrication and costly vacuum-deposition processes of organic semiconductors.

## Experimental

### Materials and methods

Commercial chemicals, which were purchased from Sigma-Aldrich, TCI, Wako Chemicals, or Kanto Chemicals, were used without further purification unless noted otherwise. All reactions were conducted under Ar in oven-dried glassware.

### Measurements

Electronic absorption spectra were recorded on a JASCO V-670 spectrophotometer. X-Ray diffraction (XRD) measurements were carried out using a Rigaku MiniFlex 600 (Ni-filtered Cu K radiation, Rigaku, Tokyo, Japan). X-Ray photoelectron spectroscopy (XPS) was performed using a PHI Quantera SXM (ULVAC-PHI, Japan) with a monochromatic Al K $\alpha$  X-ray source. Atomic force microscopy (AFM) was performed under ambient conditions with a Bruker MultiMode 8 atomic force microscope. Imaging was conducted in the ScanAsyst mode using a silicon cantilever (NCHV-A) with a frequency of 270–400 kHz. The optical texture of the organic semiconductor crystals was analyzed by polarizing optical microscope (Olympus BX51N-31P-O3, Olympus, Tokyo, Japan), equipped with a DP22 digital camera.

### Device measurements

The film thickness of each layer was measured using a microfigure measuring instrument (SURFCORDER ET200; Kosaka Laboratory, Tokyo). Devices were fabricated on highly doped *n*-type Si(100) wafers with a 300 nm-thick SiO<sub>2</sub> layer. In this



Fig. 1 (a) Schematic representation of the fabrication of OFET memory devices. (b) Chemical structures of TIPS-pentacene and polymer dielectrics of CuSP and PS<sub>4</sub>.



study, all of the results for memory performance were averaged from at least ten devices from five batches. All measurements on the transistor memory devices were conducted with a Keithley 2636B source measurement unit under dark ambient conditions. Triaxial cables were connected to the probe station to minimize background noise. The effective charge carrier mobility ( $\mu_{\text{eff}}$ ) and threshold voltage ( $V_{\text{th}}$ ) values were estimated from the slope and intercept of the linear plot of the square root of the drain-to-source current ( $I_{\text{ds}}^{1/2}$ ) vs. the gate voltage ( $V_{\text{g}}$ ) in the saturation regime using the following equation:

$$I_{\text{ds}} = WC_{\text{tot}}\mu_{\text{eff}}(V_{\text{g}} - V_{\text{t}})^2/(2L) \quad (1)$$

where  $C_{\text{tot}}$  is the capacitance per unit area of the total dielectric layer and  $L$  and  $W$  are the channel length and width, respectively.

The relations between the capacitance ( $C_{\text{tot}}$ ) of the device,  $\text{SiO}_2$  wafer ( $C_{\text{SiO}_2}$ ), polymers ( $C_{\text{poly}}$ ), and polymer dielectric constant ( $\epsilon$ ) are defined as

$$\frac{1}{C_{\text{tot}}} = \frac{1}{C_{\text{poly}}} + \frac{1}{C_{\text{SiO}_2}} \quad (2)$$

$$C_{\text{poly}} = \frac{\epsilon_0\epsilon}{d} \quad (3)$$

where  $\epsilon_0$  is the vacuum permittivity ( $8.854 \times 10^{-12} \text{ F m}^{-1}$ ),  $d$  is the thickness of the dielectric, and  $C_{\text{SiO}_2}$  is  $11.5 \text{ nF cm}^{-2}$ . The capacitances were calculated using eqn (2)–(3) in conjunction with the reported dielectric constants of 2.24 for **CuSP** and 2.48 for **PS<sub>4</sub>**, respectively.<sup>33</sup>

## Results and discussion

### Structure of blend films of organic semiconductor and polymer dielectrics

The preparation of star-shaped polystyrene with a copper phthalocyanine core (**CuSP**) and a number-average molecular weight ( $M_n$ ) of  $16.7 \text{ kg mol}^{-1}$  has been previously reported.<sup>33</sup> Four-armed polystyrene without the CuPc core (**PS<sub>4</sub>**) ( $M_n = 20.5 \text{ kg mol}^{-1}$ ) was also prepared in the present work as a reference compound. TIPS-pentacene, a derivative of pentacene, was selected as a soluble organic semiconductor, and the device performance was compared with that reported in previous studies in which vacuum-deposited pentacene was used as an organic semiconductor. The chemical structures of all the compounds studied in the present work are shown in Fig. 1b. First, **CuSP** and TIPS-pentacene were dissolved in organic solvents at a concentration of  $10 \text{ mg mL}^{-1}$ ; each solution was then mixed in a 1 : 1 volume ratio and stirred for 2 h under ambient conditions. The mixture was spin-coated at 1000 rpm onto a Si wafer substrate; the resultant films were vacuum dried to remove any remaining solvent. The film morphology was strongly influenced by the choice of solvent. Therefore, to obtain an ideal film morphology for OFET memory device applications, we tested three different solvents—chloroform ( $\text{CHCl}_3$ ), toluene, and ODCB—for preparing the films. Blend films were also prepared from a mixture of **PS<sub>4</sub>** and TIPS-pentacene for



Fig. 2 (a) XRD profiles of TIPS-pentacene/CuSP blend films spin-coated from various solvents. (b) AFM height images of the TIPS-pentacene/CuSP blend films.

comparison. The thicknesses of the TIPS-pentacene/polymer films spin-coated from  $\text{CHCl}_3$ , toluene, and ODCB were found to be approximately 100, 60, and 50 nm, respectively.

The spin-coated blend films were analyzed by X-ray diffraction (XRD) to characterize the structure of the TIPS-pentacene in the blend film. As shown in Fig. 2a, the out-of-plane XRD patterns of the mixed film obtained from ODCB exhibited strong and sharp diffraction peaks corresponding to the crystal lattice of the (00l) planes of TIPS-pentacene, indicating a crystalline structure parallel to the substrate, with an interplanar  $d$ -spacing of  $16.3 \text{ \AA}$  estimated from the peak at  $2\theta = 5.36^\circ$ . This result indicates that the triisopropylsilylethynyl groups of the TIPS-pentacene molecules align in the edge-on orientation with respect to the substrates, consistent with a single crystal of TIPS-pentacene with a domain spacing of  $16.8 \text{ \AA}$ .<sup>35,36</sup> The XRD pattern of the blend film spin-coated from toluene solution showed an additional peak at a  $d$ -spacing of  $6.92 \text{ \AA}$ , indicating that the crystals oriented in directions other than (00l) were included in the mixture. The XRD pattern of the film prepared from  $\text{CHCl}_3$  did not show clear peaks; instead, the amorphous halo originating from the polymer chains was observed at  $2\theta \approx 20^\circ$ . The surface morphology analyzed using atomic force microscopy (AFM) supported these results (Fig. 2b). The large continuous crystal grains of TIPS-pentacene were observed in the blend films spin-coated from ODCB, whereas small needle-like crystals with a size of  $\sim 0.5 \text{ \mu m}$  and a random orientation were observed in the blend film spin-coated from toluene. The film from  $\text{CHCl}_3$  exhibited a relatively smooth surface without large crystal domains. These results reveal that the crystallization of the TIPS-pentacene in the star-polymer matrix was highly dependent on the solvent, mainly because of the difference in the solvents' evaporation rate. Slow evaporation of an ODCB solution induced phase-separation of the TIPS-pentacene and star-shaped polymer, promoting crystallization of the TIPS-



pentacene, whereas fast evaporation inhibited the formation of crystals in the case of the film spin-coated from  $\text{CHCl}_3$  solution. Approximately the same XRD patterns and AFM images were obtained for the blend films prepared using  $\text{PS}_4$  as the polymer matrix, implying that the CuPc core in CuSP did not disturb the crystallization of the TIPS-pentacene (Fig. S1 and S2, ESI†).

The blend films on quartz plates were also analyzed by polarized optical microscopy (POM). As shown in Fig. 3a, large spherulite textures were observed for the blend films spin-coated from ODCB. The blend film spin-coated from a toluene solution showed smaller spherulite textures; the film spin-coated from  $\text{CHCl}_3$  did not show large crystals. The different morphologies of the TIPS-pentacene in the blend films spin-coated from the various solvents were also examined by UV-vis absorption spectroscopy (Fig. 3b). The absorption spectrum of TIPS-pentacene in a  $\text{CHCl}_3$  solution showed three characteristic absorption bands in the 500–700 nm region; these bands originated from the first excited singlet state. A similar spectrum was observed for a film spin-coated from a  $\text{CHCl}_3$  solution of TIPS-pentacene/CuSP, indicating that, like the TIPS-pentacene in the solution state, the most of the TIPS-pentacene molecules in the polymer film existed in monomeric form. The fast evaporation of  $\text{CHCl}_3$  solution during the spin-coating process might disperse the TIPS-pentacene molecules in the polymer matrix before crystallization proceeded. An additional shoulder band at  $\sim 700$  nm was observed in the spectrum of the film obtained from toluene solution, which originated from the aggregated structure of TIPS-pentacene. The spectrum of the blend film spin-coated from ODCB solution showed broader absorption bands at approximately 600, 650, and 700 nm. These results reveal that most of the TIPS-pentacene molecules assembled to form crystals even in the presence of the polymer matrix. Notably, the absorbance from the CuPc core at  $\sim 630$  nm in the spectra of the blend films was



Fig. 3 (a) POM images of the TIPS-pentacene/CuSP blend films. (b) Absorption spectra of TIPS-pentacene/CuSP blend films and TIPS-pentacene solutions.



Fig. 4 XPS depth profiles of a TIPS-P/CuSP blend films coated from (a) ODCB and (b)  $\text{CHCl}_3$  solutions on a Si substrate.

overlapped by the strong absorption of the TIPS-pentacene because of the small amount of CuPc unit in the polymer matrix.

We used XPS to investigate the detailed structure in the blend films of TIPS-pentacene and CuSP spin-coated from ODCB and  $\text{CHCl}_3$  on the Si wafer substrates. The depth profiles of the film were collected by  $\text{Ar}^+$ -ion sputtering with an etching rate of  $2.7 \text{ nm min}^{-1}$  for the film from ODCB and  $3.2 \text{ nm min}^{-1}$  for the film from a  $\text{CHCl}_3$  solution. All XPS peaks were referenced to the neutral C 1s peak at a binding energy of 284.8 eV. The peak intensities at binding energies corresponding to C 1s and Si 2p were plotted against the sputter time (Fig. 4). Notably, the Si 2p binding energies in the spectra of the TIPS-pentacene and  $\text{SiO}_2$  on the Si-wafer substrates were sufficiently different for each component to be identified (Fig. S3, ESI†). In addition, the peaks of N 1s or Cu 2p in the spectra of CuSP were difficult to observe because of the small atom%. The depth profiles of the blend film spin-coated from ODCB indicate that the TIPS-pentacene phase was located at the upper interface and mainly existed to a depth of  $\sim 20$  nm from the top interface in a blend film with a total thickness of  $\sim 50$  nm. This result is consistent with the aforementioned AFM analyses. On the other hand, the peak intensities of Si 2p corresponding to TIPS-pentacene in the  $\text{CHCl}_3$ -processed blend film were relatively constant throughout the film thickness. This result indicated that the TIPS-pentacene molecules were distributed in the polymer film without clear phase segregation, which also agreed with the above results. We concluded that the TIPS-pentacene forms large crystals on the CuSP polymer layer *via* vertical phase segregation of the blend film deposited by a simple one-pot spin-coating process from ODCB solution.

### OFET characteristics

Having successfully prepared vertically segregated organic semiconductor and polymer layers on the substrates *via* a one-pot solution process, we further fabricated OFET devices with a bottom-gate/top-contact configuration (Fig. 1a). Top-contact gold electrodes with a channel length ( $L$ ) of  $50 \mu\text{m}$  and a width ( $W$ ) of  $1000 \mu\text{m}$  were deposited onto blend films of TIPS-pentacene and star polymers of CuSP or  $\text{PS}_4$  spin-coated onto highly doped n-type Si(100) wafers with a 300 nm-thick surface layer of  $\text{SiO}_2$ . The capacitance of the gate dielectric ( $C_{\text{top}}$ ) was estimated on the basis of the reported dielectric constants







Fig. 7 Retention time of the  $I_d$  of the OFET memory device with TIPS-pentacene/CuSP blend films, as monitored at  $V_g = -5$  V after writing (black squares) and erasing (white circles) operations.

multiple switching durability of the OFET with CuSP was further evaluated through write-read-erase-read (WRER) cycle test. The drain current was read at  $V_g = -10$  V with the fixed  $V_d = -50$  V after repeating the writing and erasing operations with the continuous bias pluses of  $-50$  V/50 V (Fig. 6a). After several cycles, the  $I_{ON}/I_{OFF}$  ratio was maintained above  $10^2$  over 100 cycles (Fig. 6b).

Charge retention is also important when evaluating the nonvolatility of a memory device. To evaluate the memory retention characteristics, we monitored the drain current at  $V_g = -5$  V after a writing operation at  $V_g = -50$  V for 1 s and an erasing operation at  $V_g = +50$  V for 1 s (Fig. 7). The difference in the drain on/off current was maintained at  $10^2$  for 12 000 s. High stability of the stored charge was achieved because of the unique architecture of the star-shaped polymer, where the arm polymer chains surrounding the CuPc core assemblies restrain the leakage of the trapped charges.

## Conclusions

We have successfully developed high-performance solution-processed OFET memory devices using TIPS-pentacene and copper phthalocyanine-cored star-shaped polystyrene. The morphology and crystal structure of the TIPS-pentacene in the polymer blend film was found to be dependent on the solvent. We also found that the TIPS-pentacene/polymer film spin-coated from ODCB solution showed vertical phase separation, with large crystal grains of the organic semiconductor at the top surface, which is advantageous for OFET device applications. The OFET devices fabricated by the solution process using the blend films of TIPS-pentacene and CuSP exhibited a high charge carrier mobility with substantial memory characteristics. The novel methodology of using a polymer nano-floating gate to construct OFET memory in a one-pot solution process is a promising technique to prepare high-performance OFET memory devices and would be useful for fabricating flexible and large-area memory devices.

## Conflicts of interest

There are no conflicts to declare.

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