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Soluble Fullerene-Based n-Channel Organic Thin-Film Transistors Printed by Using a Polydimethylsiloxane Stamp

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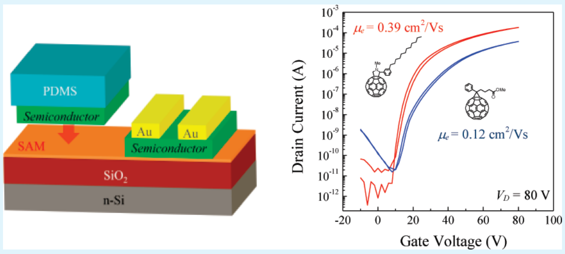

ABSTRACT: A polydimethylsiloxane stamp was applied for the first time to the fabrication of n-channel thin-film transistors based on soluble small molecule organic semiconducting materials. The stamping method was found to facilitate film transfer onto a gate insulator surface irrespective of its surface free energy. We used [6,6]-phenyl-C$_{61}$-butyric acid methyl ester (PCBM) and C$_{60}$-fused *N*-methylpyrrolidine-*meta*-dodecyl phenyl (C60MC12) as n-channel materials. The stamped thin-film transistors of C60MC12 achieved a high electron mobility of 0.39 cm^2/(V s) and a current on−off ratio of 1×10^7. The mobility of the stamped C60MC12 thin-film transistors did not depend much on the surface free energy of the SiO$_2$ gate insulator with and without surface treatment using a silane-coupling reagent. In particular, the stamped C60MC12 thin-film transistor exhibited a relatively high mobility of 0.1 cm^2/(V s) on a high energy surface of untreated SiO$_2$. In addition, a complementary inverter composed of an n-channel and a p-channel stamped thin-film transistor was demonstrated for the first time, which exhibits a maximum gain of 63 at a supply voltage of 50 V.

KEYWORDS: organic field-effect transistor, organic semiconductor, n-channel material, soluble fullerene derivative, polydimethylsiloxane stamp, transfer printing, self-assembled monolayer, complementary inverter

1. INTRODUCTION

Organic thin-film transistors (TFTs) have been vigorously investigated for various applications to "plastic electronics" such as flexible displays, radio frequency identification tags, sensors.[1−9] In recent years, several attempts have been reported on more integrated devices including organic complementary circuits, a power-saving logic circuit combining p- and n-channel organic TFTs. Development of fine patterning and environment-friendly fabrication techniques are required for applying organic TFTs to various practical devices.

Printing techniques will provide energy-saving and low-cost fabrication processes for these organic electronic devices.[10] Screen printing[11,12] and inkjet printing[13,14] have been often examined for fabrications of organic devices. Recently, microcontact printing (μCP) also started to be employed as a high-resolution patterning method. It is a transfer printing technique first demonstrated by Whitesides et al. for transferring alkylthiols on a gold film,[15] and can provide much finer patterns of submicrometer order than conventional printing techniques. An elastomeric stamp out of polydimethylsiloxane (PDMS) was used to transfer inks. The feature of the transfer printing including μCP is that an ink is transferred from a surface with low surface free energy to an objective surface. Several groups have reported μCP for electrode and semiconductor fabrication of organic TFTs.[1−3,5,6,16−29] However, organic semiconductors so far examined are limited to p-channel polymers such as poly(3-hexylthiophene) (P3HT) and poly(3,3‴-dialkylquaterthiophene).[6,20,29] The applicability of μCP for other semiconducting materials, especially nonpolymer materials, has not been sufficiently investigated. In addition, transfer printing with a PDMS stamp is an effective method to bring out a better performance of organic TFTs of crystalline organic materials. Takakuwa et al. reported improved crystallinity and TFT performances of "stamped" P3HT films compared with spin-coated films.[29]

This paper is the first report for transferred films of soluble small molecule semiconducting materials and their n-channel TFTs by PDMS stamping.[30] We used fullerene derivatives, [6,6]-phenyl-C$_{61}$-butyric acid methyl ester (PCBM) and C$_{60}$-fused *N*-methylpyrrolidine-*meta*-C12 phenyl (C60MC12) (Figure 1a) as inks for the PDMS stamping. PCBM is a frequently used n-channel material for organic electronics. C60MC12 is one of the soluble n-channel materials we developed for organic TFT application.[31−33] It shows high solubility for organic solvents and forms an ordered crystalline film by spin-coating. The TFT devices based on C60MC12 exhibited a high mobility of about 0.5 cm^2/(V s) under the optimal surface treatment of their gate insulator, which is one of the highest values for n-channel organic TFTs with soluble semiconducting materials.[34]

This paper also clarifies the detailed structure of "stamped" films and its effects on TFT performances in comparison with spin-coated films. The structural features of a semiconductor-gate insulator interface, such as surface roughness, molecular packing and domain structure of a semiconductor layer, and the number of

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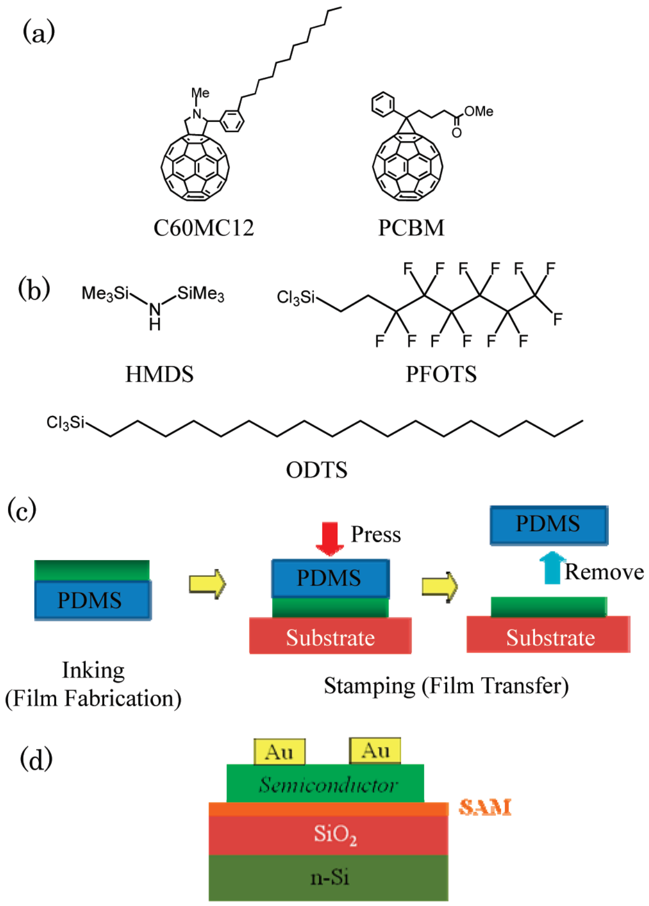

Figure 1. (a) Soluble n-channel materials. (b) SAM materials. (c) Film fabrication and transfer process of the PDMS stamping. Green part represents a film. (d) Schematic of a stamped TFT.

carrier traps, has a strong effect on TFT performances, since the majority of charge carriers in a channel transports in the vicinity of this interface. For example, self-assembled monolayer (SAM) treatment of a gate insulator surface often improves electronic performances of bottom-gate type organic TFTs.[34−39] However, independent estimation of more than one possible factor is not always easy, because both film crystallinity and the number of carrier traps are often affected at the same time by SAM treatment. Transfer technique should simplify this problem by eliminating the film crystallinity factor; as the surface of a semiconductor film is already dry on a PDMS stamp, the structure of the "stamped" film should be dependent only on the nature of PDMS, i.e., its first substrate, and should not depend much on the gate insulator surfaces where it is transferred afterward.

Moreover, this paper demonstrates a successful thin film fabrication on a hydrophobic surface by optimizing transfer condition of ink. Generally, a hydrophobic surface tends to shed a solvent, which makes film fabrication difficult by conventional wet processes such as spin-coating, inkjet printing, and screen printing. The PDMS stamping avoids this difficulty and makes films with a good reproducibility. Furthermore, PDMS stamps enable us to fabricate various patterned films composed of different materials. We therefore demonstrate an organic complementary inverter based on P3HT and C60MC12 TFTs on the same substrate.

2. EXPERIMENTAL SECTION

2.1. Materials. C60MC12 was synthesized via a similar route as that in the previous reports.[32,40] PCBM and P3HT were purchased from Frontier Carbon Corporation and Merck, respectively, and used without further purification. Three types of silane-coupling reagents, hexamethyldisilazane (HMDS), octadecyltrichlorosilane (ODTS), and 1H, 1H, 2H, 2H-perfluorooctyltrichlorosilane (PFOTS) (Figure 1b) were purchased from Aldrich, and used without further purification. A highly doped n-type silicon wafer covered with 300 nm thick SiO_2 was used as a substrate as well as a gate and a gate insulator (a capacitance per unit area of 10 nF/cm^2).

2.2. SAM Treatments. The SiO_2 surface of a silicon wafer was cleaned sequentially with acetone, an alkaline solution (Furuuchi Chemical Semico Clean 56), ultrapure water, and ethanol in an ultrasonic bath and afterward with ultraviolet/ozone. The substrate was then treated with the silane-coupling reagents by soaking it in either a neat liquid of HMDS, a 0.1 M toluene solution of ODTS, or a 0.01 M toluene solution of PFOTS for 2 h. The contact angles of the gate insulator surfaces and the stamp surface for water and hexadecane were measured using a contact angle meter (Kyowa Interface Science DM-500). Then, the surface free energies were calculated by means of the Kaelble-Uy method.

2.3. Fabrication of Semiconductor Films. A 10 g of PDMS precursor (Shin-Etsu KE-106) and a 1 g of cross-linker (Shin-Etsu CAT-RG) were mixed, deaerated carefully, and poured onto a glass plate whose surface was pretreated with fluorinated SAM to ensure facile separation. Another glass plate pretreated with SAM was then placed onto the poured material. After 30 min of cure at 150 °C, the resulting PDMS stamp together with the upper glass plate was peeled off from the lower glass plate. The semiconductor films were fabricated on the SiO_2 gate insulator surfaces with and without SAM treatments by means of film-transfer (Figure 1c). The PDMS stamp was cleaned with acetone by spin-casting and the static charge on the stamp surface was eliminated with an ionizer (Shishido Electrostatic WINSTAT BF-22A). The semiconductor films were formed on the PDMS plate by spin-coating a 10 mg/mL chloroform solution of a semiconductor material. Spin-coating condition was 2,000 rpm for 6 s. Then, the films were immediately transferred to the gate insulator surfaces; the PDMS stamp was turned upside down, attached on the substrate, left for a while, and peeled off from the semiconductor film. The substrate was heated at 50 °C during the stamping. The PDMS stamps stand repetitive use. In the case of direct spin-coating on the SAM-treated SiO_2 gate insulators, the hydrophobic insulator surfaces were edged with a hydrophilic pattern of 30-nm-thick evaporated SiO to ensure uniform film fabrication.[34] The thickness of the stamped and spin-coated semiconductor films is 50−60 nm.

2.4. Device Fabrications and Measurements. The TFT structure is the bottom-gate-top-contact type (Figure 1d). A 30 nm thick layer of gold was vacuum-evaporated on the stamped and spin-coated films as source and drain electrodes, and the TFTs were transferred to a vacuum chamber (Thermal Block Company LN2 prober system) without exposing to air. After annealing the film at 100 °C for 12 h in a vacuum, the TFT characteristic measurements were performed by means of a semiconductor parameter analyzer (Keithley 4200-SCS) at room temperature in a vacuum (5×10^{-6} Torr). The field-effect mobilities are extracted from the saturated regimes using the equation as follows: $I_D = (WC/2L)\mu\ (V_G - V_T)^2$, where μ is the field-effect mobility, I_D is the drain current, W (1 mm) is the channel width, L (40 μm) is the channel length, C (10 nF/cm^2) is the capacitance per unit area of the insulator, and V_G and V_T are the gate voltage and the extrapolated threshold voltage, respectively. X-ray diffraction (XRD) measurements of the thin films were carried out on a Rigaku Denki RU-300 using Cu Kα radiation (40 kV, 200 mA). The diffractions were measured from 2 to 30° in the $2\theta-\theta$ scan mode with a 0.01° step. Noncontact mode atomic

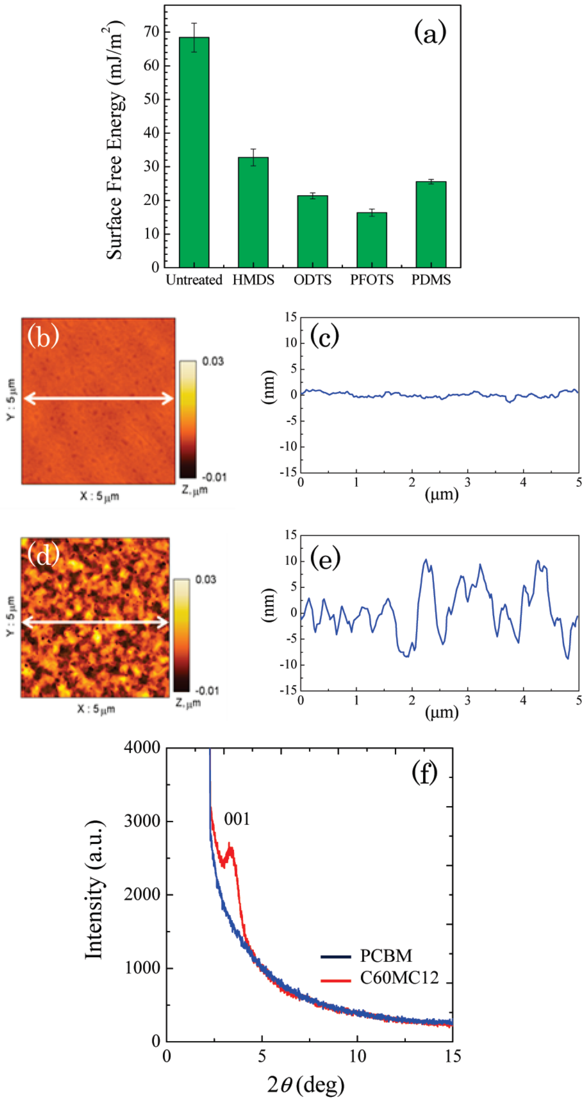

Figure 2. (a) Surface free energies of the SAM-treated gate insulators and PDMS. (b) AFM topview and (c) cross-sectional profiles of the PCBM stamped film, (d) AFM topview and (e) cross-sectional profiles of the C60MC12 stamped film, and (f) XRD patterns of the stamped films. Blue and red lines correspond with PCBM and C60MC12, respectively, on the ODTS-treated SiO_2 insulator.

force microscopy (AFM) was performed on an SII Nanotechnology SPA-300 equipped with an SPI3800 controller for morphological observations of the films. A complementary inverter circuit was fabricated by stamping both a p-channel (P3HT) and an n-channel (C60MC12) film on a same substrate. Gold source and drain electrodes with the same dimension as above were vacuum-evaporated, and the p-channel and the n-channel TFTs were electrically connected with gold wires. The circuit was again set to the vacuum chamber for inverter measurements using two computer-controlled source measurement units (Keithley 6430 and 2400 sourcemeters) and an electrometer unit (Keithley 6517A).

3. RESULTS AND DISCUSSION

3.1. Surface Free Energy and PDMS Stamping. Figure 2a shows the surface free energies of various SAM-treated SiO_2. The relation of the surface free energy was as follows: untreated > HMDS > ODTS > PFOTS. The surface free energy of the PDMS stamps was 25.6 mJ/m^2, which is higher than those of the ODTS- and the PFOTS-treated surfaces.

Generally, film transfer to a device substrate occurs when the surface free energy of the device substrate is higher than that of an original substrate.[41] According to the above results, film transfer from the PDMS stamp to the ODTS- and PFOTS-treated surfaces was expected to be difficult. However, the semiconductor films were successfully transferred to all the untreated and SAM-treated gate insulator surfaces including these lower energy surfaces than the PDMS surface. We presume the following transfer mechanism: (1) a spin-coated solution of a semiconductor on a PDMS stamp should start drying from the air—solution interface rather than the solution-PDMS interface. (2) As a result, the solution-PDMS interface can be still wet while the surface of the semiconductor film is already dry, which may allow film release from the PDMS stamp. In addition, a small volume change of PDMS by swelling with a solvent may help facile film transfer. The fact that a fully dried film on a PDMS stamp was hardly transferred to the lower energy surfaces also supports the above assumption. Furthermore, the transferability was improved by heating the substrate during the film transfer, which may be related to a change of the surface free energy of PDMS and a solvent evaporation rate from the spin-coated film.

The above results also show that the stamping method can be applied not only to polymer semiconductors but also soluble small molecule materials. However, not every material is available for fabricating a uniform film with sufficient thickness; films of several materials including 6,13-bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) were not well-transferred by this method. Such materials tend to be absorbed by a PDMS stamp together with their solvent, which we presume is a cause for their ill transfer. This may be related to the molecular size of each material: C60MC12 ($C_{81}H_{25}N_1$ = 1012.07), PCBM ($C_{72}H_{14}O_2$ = 910.88), and TIPS-pentacene ($C_{44}H_{54}Si_2$ = 639.07). On the other hand, it is unlikely the case that the stamping process depends on a speed of aggregation of each material during the spin-coating process on a PDMS stamp, because PCBM and C60MC12 have comparable solubilities to that of TIPS-pentacene.

3.2. Structural Aspect of the Stamped Films. The surface morphology observed by AFM was different for PCBM (Figure 2b,c) and C60MC12 (Figure 2d,e). The PCBM film has a very smooth surface. On the other hand, the C60MC12 film has a rugged surface with a height difference of 10 nm, which suggests crystallized grains. These tendencies are similar to those of their spin-coated films.[31]

Figure 2f shows the XRD patterns of the stamped films on the ODTS-treated SiO_2 surface. We observed no diffraction peaks for any of the PCBM films irrespective of the SAM treatment, suggesting its amorphous nature.[31] On the other hand, a 001 diffraction peak was observed for all the C60MC12 films irrespective of the SAM treatment, suggesting that the stamping affords a crystalline film as well as those by spin-coating.[31,42] Moreover, the peak intensity and the peak width of the 001 diffraction were independent of the SAM treatment, i.e., the crystallinity of the C60MC12 stamped films is independent of the SAM treatment. This result is in contrast to the case of the spin-coated C60MC12 films, where clear dependence in crystallinity on the SAM treatment was observed.[34]

The above results show the molecular packing of both of PCBM (amorphous) and C60MC12 (crystalline) in the stamped films is independent of SAM treatment. This is in accordance with our assumption that a stamped film transferred onto a gate

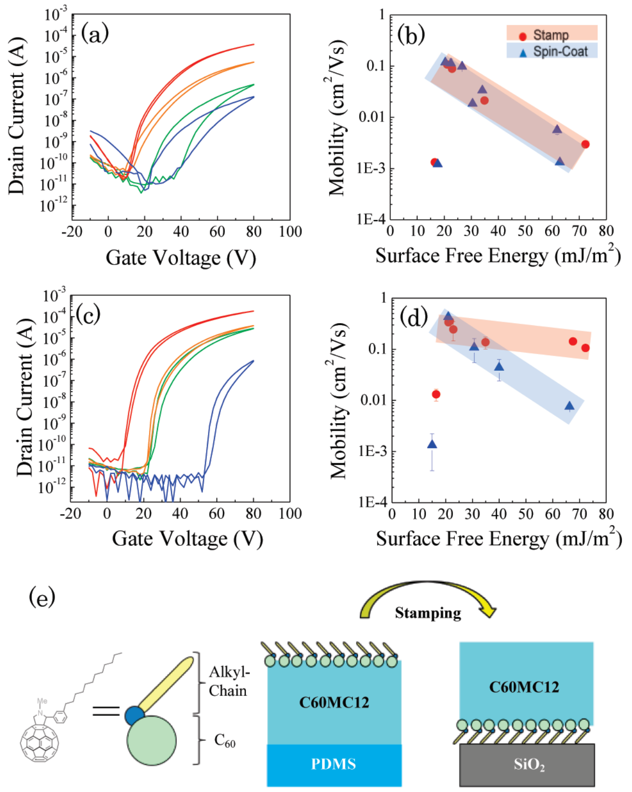

Figure 3. Transfer characteristics of (a) the PCBM stamped TFTs and (c) the C60MC12 stamped TFTs at a constant source-drain voltage of 80 V. The green, yellow, red, and blue lines correspond with the untreated, HMDS-treated, ODTS-treated, and PFOTS-treated insulators, respectively. Relation between charge mobility and surface free energy of the insulator for (b) the PCBM TFTs and (d) the C60MC12 TFTs. Red circles and blue triangles represent the results for stamping and spin-coating, respectively. (e) Models of the film transfer process and the transferred film of C60MC12 with ordered alkyl chains at the interface.

insulator should preserve its initial packing structure formed when it starts drying on a PDMS stamp.

3.3. n-Channel Stamped TFTs. Figure 3a shows the transfer characteristics of the stamped TFTs of PCBM. A large hysteresis was observed for the devices with the untreated SiO_2 insulator and the PFOTS-treated one. On the other hand, the devices with the HMDS- and ODTS-treated insulators exhibited only a small hysteresis, and their drain current turned on sharply compared with the former. Figure 3b shows the relation between the mobility of the PCBM TFTs and the surface free energy of the insulators depending on the fabrication method of the film. The stamped films and the spin-coated films showed similar dependency of mobility on the surface free energy; the lower the surface free energy of the insulator down to 20 mJ/m^2, the higher the mobility of the PCBM TFT. The improvement of the mobility for the PCBM TFTs at the lower surface energy region is dominated by reduction of an interference, chemically and/or spatially, of carrier trap species (silanol groups) on the SiO_2 insulator surface by alkyl-SAM treatment ("the passivation effect")[36,37] rather than molecular ordering at the channel region of the semiconducting layer ("the structural effect"),[38,39] because as the XRD results show, the PCBM films are amorphous on all of the SiO_2 insulators irrespective of their surface free energies. The possibility is not completely excluded, however, that the molecular order of PCBM at the PCBM-insulator interface is improved by SAM treatment, considering the fact that PCBM is reported to take an ordered molecular packing under a certain condition.[43−46]

Figure 3c shows the transfer characteristics of the stamped TFTs of C60MC12. All of the C60MC12 TFTs showed a sharp turn-on with only a small hysteresis, irrespective of the SAM treatment on the insulator.[47] The ODTS-treated TFT showed the largest on-current and the smallest turn-on voltage. Figure 3d shows the relation between the mobility of the C60MC12 TFTs and the surface free energy of the insulators depending on the fabrication method of the film. The highest mobility values were obtained in the region of surface free energy around 20 mJ/m^2 for both of the spin-coated and the stamped TFTs. However, the stamped TFTs maintained relatively high mobilities even in the high energy region (Figure 3d, red line), which is in contrast to the large dependency observed for the spin-coated TFTs (Figure 3d, blue line).

The effect of SAM treatment on the mobility for the spin-coated C60MC12 TFT was interpreted by both of the two effects, "the structural effect" and "the passivation effect".[34] On the other hand, "the structural effect" can be almost excluded for the stamped C60MC12 TFTs, since the crystallinity of all the films are similar irrespective of the SAM treatment. The relatively high mobility of the stamped C60MC12 TFTs regardless of the surface free energy of the insulator surface suggests that the C60MC12 film is "well-passivated" from silanol groups even on the untreated SiO_2 surface. Considering the film formation process by stamping, we propose a model of the stamped film of C60MC12 as illustrated in Figure 3e. (1) The C60MC12 solution is spin-coated onto the PDMS stamp. (2) When being dried, C60MC12 faces its hydrophobic alkyl chains to the air at the surface of the spread solution. (3) When transferred onto the gate insulator, the outermost alkyl chains should contact to the insulator surface and separate the fullerene portion from the insulator surface. This may reduce the interference of silanol groups on the SiO_2 surface that can act as electron traps, especially when the film is transferred onto the untreated SiO_2 surface. The above experimental results and our assumption also indicate a possibility that a combination of stamping technique and an organic semiconductor material with long alkyl chains may provide a high-performance TFT without being influenced by the nature of an insulator surface.

Table 1 summarizes the TFT characteristics of PCBM and C60MC12. ODTS treatment afforded the best performance for both of the materials. The best mobilities for the stamped PCBM and C60MC12 TFTs are 0.12 and 0.39 $cm^2/(V\ s)$, respectively, with on/off ratios of more than 1×10^6. The C60MC12 TFTs exhibit a higher mobility than the corresponding PCBM TFTs, which is due to the higher film crystallinity of the former.[31] Even though the best mobility of the stamped C60MC12 TFTs did not reach that of the spin-coated devices, their TFT performances are already comparable with those of the spin-coated films, and will be further improved by fabricating a more ordered molecular packing on the PDMS stamp.

3.4. Stamped Complementary Inverter. The stamping enables film fabrication of more than one different material on a same substrate. A complementary inverter based on a P3HT and a C60MC12 transistor was fabricated by PDMS stamping, where the films of the both materials are on a same substrate (Figure 4a,b). The inverter characteristics exhibit a high gain of 63 and a turnover point of ca. 34 V (Figure 4c,d), which is as good as the spin-coated complementary circuit composed with separated substrates of the p- and n-channel TFTs of P3HT and C60MC12 (a gain of 68, data not

Table 1. **Summary of n-Channel TFT Characteristics for Soluble Fullerene Derivatives**

material	method	SAM	μ (cm^2/(V s))	I_{ON}/I_{OFF}	V_{TH} (V)	S (V/dec)
C60MC12	stamp	PFOTS	0.01	3.3×10^5	59	1.3
C60MC12	stamp	untreated	0.10	1.3×10^7	32	1.3
C60MC12	stamp	HMDS	0.14	9.8×10^6	32	1.1
C60MC12	stamp	ODTS	0.39	4.1×10^7	18	1.2
C60MC12	spin-coat	ODTS	0.45	5.2×10^7	24	1.1
PCBM	stamp	PFOTS	6×10^{-4}	3.2×10^3	38	2.1
PCBM	stamp	untreated	0.002	1.3×10^5	40	3.6
PCBM	stamp	HMDS	0.02	3.0×10^5	32	3.4
PCBM	stamp	ODTS	0.12	2.4×10^6	29	2.3
PCBM	spin-coat	ODTS	0.13	5.9×10^6	25	2.1

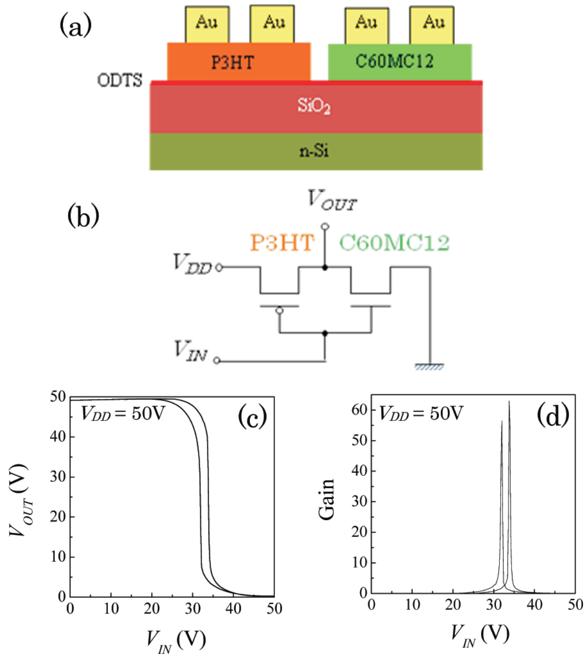

Figure 4. (a) Schematic of the P3HT/C60MC12 stamped inverter. (b) Circuit diagram of the inverter. (c) Transfer characteristic of the P3HT/C60MC12 stamped inverter and (d) gain characteristic from the transfer curve of the P3HT/C60MC12 stamped inverter with a supply voltage (V_{DD}) of 50 V.

shown). These gains are relatively high compared with the previous reports on the inverters based on spin-coated films of PCBM:P3HT mixture,[48] spin-casted films of polytriarylamine (PTAA) and PCBM,[49] gravure printed films of p- and n-channel polymers.[50] Performance of the stamped complementary inverters still does not exceed the vacuum-deposited organic complementary inverters with a gain greater than 100.[51−55] The gain of stamped inverters will be improved by selecting an appropriate p-channel semiconducting material. The relatively high turnover point of the present circuit can be adjusted by tuning the channel width, the channel length, and so on.

4. CONCLUSIONS

A good film transfer property covers a surface free energy from 17 to 72 mJ/m^2. One of the advantages of PDMS stamping is printability on an insulator surface irrespective of its surface free energy. The correlation between the mobility and the surface free energy were investigated for the stamped TFTs. The devices exhibited higher charge mobility on the insulator surface with lower surface free energy in the range from 20 to 70 mJ/m^2. However, the C60MC12 TFTs fabricated by spin-coating and stamping exhibited different mobility dependencies on the surface free energy of the gate insulator surfaces. The stamped TFT of C60MC12 showed a high mobility of 0.1 cm^2/(V s) even on the high energy surface. This suggests that the structure of the semiconductor-gate insulator interface depends on the film fabrication methods. The above results demonstrate the applicability of the stamping method for TFT fabrication, especially with n-channel organic semiconductors with long alkyl chains. The complementary P3HT/C60MC12 inverter fabricated by PDMS stamping exhibited a high gain of 63.

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