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Functionalized Arylacetylene Oligomers for Organic Thin-Film Transistors (OTFTs)**

By V. A. L. Roy, Yong-Gang Zhi, Zong-Xiang Xu, Sze-Chit Yu, Philip Wai Hong Chan, and Chi-Ming Che*

The search for new classes of organic and metal-organic compounds for organic thin-film transistors (OTFTs) is of immense current interest due to the relatively low cost and potential applications of OTFTs in flexible large-area electronic devices such as displays^[1,2] and sensors.^[3–5] Indeed, OTFTs have been recently reported in the fabrication of ac-

tive-matrix displays^[6] and integrated circuits (ICs) for logic and memory chips.^[7] Unlike their silicon-based TFTs counterparts which require expensive and complicated fabricating systems at a high temperature,^[8] the deposition of organic materials on large-area substrates, including flexible polymer substrates, is much less complicated.^[1]

Over the last decade, much attention has been focused on the study of pentacene and its derivatives as organic semiconductors for OTFTs. This class of materials has been shown to exhibit excellent field-effect mobilities^[9] and environmental stabilities.^[1] Similarly, oligothiophenes,^[10] poly(3-alkyl-thiophene) (P3HT),^[11] and fused heterocyclic compounds^[12] have been shown to possess comparable properties that are highly desirable for OTFT applications. Despite these advances, difficulties in structural modification of these literature-reported organic semiconductors remain. Current methods used to modify charge-carrier properties of the materials rely on changing the type of substitution on the materials. By introducing electron-donating (D) or electron-accepting (A) functional groups, the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) levels of the materials can be fine-tuned to give either p-type or n-type organic semiconductors. In this context, it would be desirable to develop new classes of organic materials which possess extended π -conjugated electronic systems that can be easily prepared and modified and exhibit effective intermolecular interactions for charge transportation.

Work in our laboratory and by others has demonstrated the potential applications of conjugated ethynylated arylacetylene materials such as arylacetylenes,^[13] poly(arylene ethylene)s,^[14] and metal-containing acetylide complexes.^[15] These materials have received considerable attention in organic light-emitting diodes,^[16] optoelectronics,^[17] molecular electronics,^[18] and sensors.^[19] Through incorporation of different electron-donating or electron-withdrawing functional groups, the physical and electronic behavior of oligomeric arylacetylenes such as thermal stabilities, carrier transport, and structural packing properties could be readily and systematically modified for OTFT applications. Also, the π -conjugation length of the linear molecules can easily be extended by synthetically controlling the number of repeating arylacetylene moieties incorporated into the structures. For example, Wong et al. recently reported the synthesis of crystalline pyrimidine-containing arylethynyl molecules with dipole–dipole and face-to-face π – π interactions.^[20] Indeed, the relative planar configuration of arylacetylene moieties along the molecules was reported to be important for the on–off switch stage.^[21]

Herein, we introduce arylacetylene oligomers **1–3** comprising of three repeating phenylene ethynylene units containing *para*-substituted electron-donating or electron-withdrawing substituents (Fig. 1). Compounds **1–3** were prepared with product yields up to 91 % using a synthetic strategy based on a simple iterative and convergent Pd/Cu-coupling and desilylation protocol. This involved treatment of the respective trialkylsilyl iodide and terminal arylalkyne starting materials in the presence of PdCl₂(PPh₃)₂ and CuI at room temperature

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** This work is supported by the University Development Fund (Nanotechnology Research Institute, 00600009) of The University of Hong Kong. We acknowledge Sunic System Ltd. for support of the fabrication system at The University of Hong Kong.

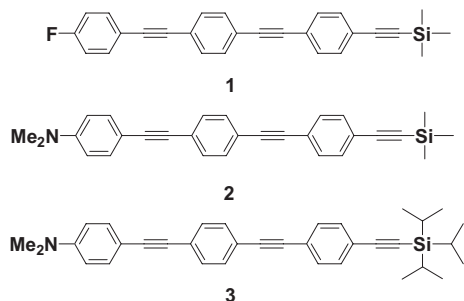


Figure 1. Structures of arylacetylene oligomers 1–3.

under Sonogashira reaction conditions.^[22] The terminal arylalkynes were prepared from their trimethylsilane-protected precursors by base-mediated desilylation. All the oligomers were purified by sublimation under high vacuum. To our knowledge, the oligo(arylacetylene)s studied in this work are the first linear rod-like molecules used for OTFT applications.

The structural order and quality of the thin films play important roles in the device performance. Atomic force microscopy (AFM) was used to examine the film surface morphology of oligo(arylacetylene)s. All thin films were prepared by deposition of the compounds on SiO₂. Among the samples, **2** showed the best surface morphology, with a surface roughness of 1.6 nm (root-mean-square) in a 1 $\mu\text{m} \times 1 \mu\text{m}$ scan area, as depicted in Figure 2. The surface morphology was smooth, and polycrystalline grain structures were observed. The morphological smoothness and formation of grains across the film reveal that only limited numbers of defects were introduced into the layer. Thermal stabilities of **1–3** were characterized by thermogravimetric analysis (TGA). The decomposition temperature (T_d) was measured with a scanning rate of 15 $^{\circ}\text{C min}^{-1}$ under a nitrogen atmosphere. The results showed that oligo(arylacetylene)s **1–3** were thermally stable, with $T_d = 281 \text{ }^{\circ}\text{C}$ for **1**, 255 $^{\circ}\text{C}$ for **2**, and 315 $^{\circ}\text{C}$ for **3**.

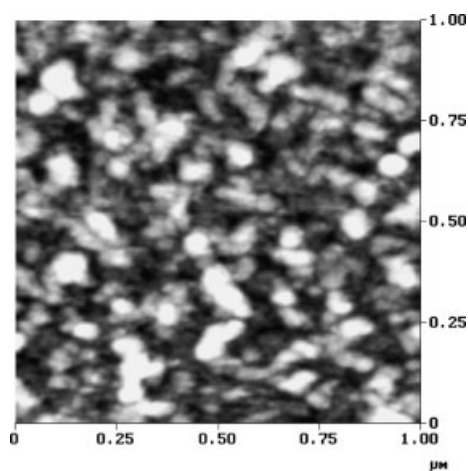


Figure 2. AFM image of **2** on SiO₂ at room temperature (5 nm average thickness).

Bottom-contact OTFT devices comprising the oligo(arylacetylene)s **1–3** as active layers were fabricated. Figure 3 shows a typical bottom-contact layout of the field-effect transistor (FET). All devices were fabricated by evaporation of

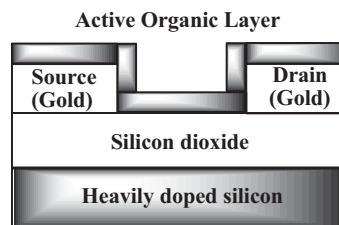


Figure 3. Schematic structure of bottom-contact OTFT.

the contact pads formed by a Ti/W adhesion layer (10 nm thick), followed by deposition of Au (100 nm) on SiO₂ layers (100 nm thick, permittivity = 3.9) which were thermally grown on n-type Si substrates (gate electrode). The source and drain electrodes were obtained by sputtering the Au layer (50 nm thick) on silicon dioxide. Patterning of the devices was performed using a lithographic process. The Au electrodes with no adhesion layer were utilized to ensure charge injection from Au and to prevent shadow effects during sublimation of the organic film layers. The profile of the Au electrode was characterized by AFM. The profile was observed to have a smooth slope and a regular pattern along the entire channel width. The active organic layer with a thickness of 50 nm was deposited with a deposition rate of 2 \AA s^{-1} on top of the patterned substrates under high-vacuum conditions.

Figures 4a,b show the plots of output characteristics (drain-source current I_{DS} versus drain-source voltage V_{DS}) and transfer characteristics (drain-source current versus gate-source voltage V_{GS}) of device A using **2** as the organic active layer. The transfer characteristics were measured at drain voltages of -10 V . Device A was characterized with a channel length and channel width of 100 μm and 1080 μm , respectively. The field-effect mobility of **2** was 0.3 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ measured at a gate-source voltage of -8 V . An on/off current ratio of 10^5 was obtained. In comparison to the OTFT comprising **3** as an active layer (device B, field-effect mobility = $4.3 \times 10^{-4} \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$), device A demonstrated a better device performance. This observation is consistent with AFM measurements revealing **2**, with a less bulky trimethylsilyl group, results in more effective structural ordering, which we surmise would provide better π -orbital interactions and more efficient charge transport. In comparison, the transistor fabricated with **1** as the active layer did not exhibit prominent field-effect behavior. In general, top-contact devices should show better performance than their bottom-contact counterparts due to the good contact of organic films at the interface of metal and organic layers in the former. The aforementioned results were cross-examined by a top-contact device, where a 50 nm thick gold electrode was deposited on the layer

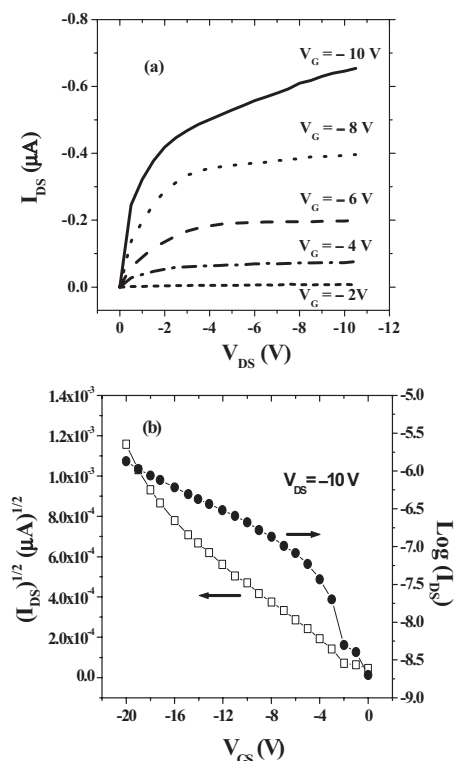


Figure 4. a) Output characteristics and b) transfer characteristics of device A.

of **2**, and similar device performance was obtained. We consider that the similar performance of top- and bottom-contact devices could be due to better interactions and matched work-function levels between the layer of **2** and the Au electrode, facilitating charge injection at the metal/organic interface.

The current-decay properties and stability of device A under prolonged bias was verified by transient measurements with a constant applied voltage to the gate and drain. Figure 5 depicts the characteristics of current decay as a function of time. Notably, the current was found to decay slowly in a non-single-exponential decay manner. In addition, the device re-

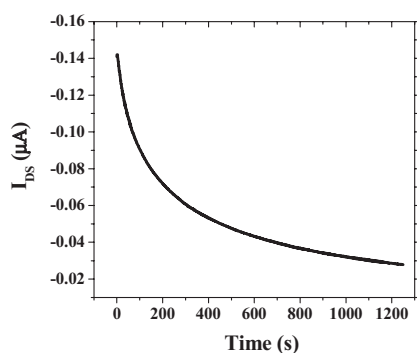


Figure 5. Transient measurements of device A at $V_{GS} = V_{DS} = -10$ V.

quired several minutes for complete current decay down to the noise threshold; this decay could be attributed to charge trapping in the device. The presence of trap states has been previously reported in amorphous silicon TFTs^[23] prior to the investigations of organic and polymeric TFTs.^[24] However, it should be noted that the nature and exact location of charge-trap states remain a matter of debate for OTFTs.

In summary, oligo(arylacetylene)s with *para*-substituted electron-donating or electron-withdrawing substituents were prepared by employing a simple Pd-catalyzed coupling strategy. This class of materials with extended π -conjugation provides a new impetus to fine-tuning HOMO and LUMO energy levels to achieve either p-type or n-type organic semiconductors by simple modification of the molecular structure. The oligo(arylacetylene)-based OTFT fabricated with good structural order and smooth film morphology showed a field-effect mobility of $0.3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. To our knowledge, this is one of the highest field-effect mobilities reported for a OTFT device that does not possess a self-assembled-layer configuration. The transistor showed a long current-decay time, revealing good device stability under prolonged operation. We envision that the field-effect mobility could be further improved through modification of the oligo(arylacetylene)s or by self-assembly of monolayers onto the device.

Experimental

Compounds **1–3** were prepared following modification of literature methods [22]. Preparation of **2** is described as a typical procedure: To a tetrahydrofuran (THF, 50 mL) solution containing [4-(4-iodophenylethynyl)-phenylethynyl]trimethylsilane (1.0 mmol), 4-ethynylphenyl-N,N-dimethylamine (1.1 mmol), and Et_2NH (10 mL) degassed with Ar for 30 min was sequentially added CuI (0.02 mmol) and $\text{PdCl}_2(\text{PPh}_3)_2$ (0.04 mmol). The resultant reaction mixture was stirred for 24 h. Upon completion, the solvent was removed and water (40 mL) was added. The mixture was extracted with Et_2O (3×30 mL) and the combined organic phases dried over MgSO_4 . Filtration, concentration in vacuo, and purification by column chromatography (8:1 *n*-hexane/ EtOAc as eluent) gave the title compound. Terminal aryl-alkynes were prepared as follows: To a THF and MeOH (1:1 v/v, 30 mL) solution containing the silyl phenylacetylene adduct (1.1 mmol) was added K_2CO_3 (16 mmol) with stirring for 10 min. The reaction mixture was subsequently allowed to stir for 8 h at room temperature. Upon completion, the solvent was removed and the residue obtained was diluted with water (30 mL) and extracted with Et_2O (2×30 mL). The organic phase was further washed with water (2×30 mL), dried over MgSO_4 , filtered, and concentrated to dryness to give the desilylated terminal alkyne.

4-[4-(4-Fluorophenylethynyl)phenylethynyl]phenylethynyl trimethylsilane (1): Yield: 91 %. Anal. Calcd. for $\text{C}_{27}\text{H}_{21}\text{FSi}$: C, 82.61; H, 5.39. Found: C, 82.15; H, 5.37. ^1H NMR (400 MHz, CDCl_3): δ 7.54–7.45 (m, 10H), 7.06 (t, $J = 8.7$ Hz, 2H), 0.26 (s, 9H). ^{13}C NMR (75 MHz, CDCl_3): δ 164.2, 160.9, 137.5, 133.5, 133.4, 133.0, 132.4, 131.9, 131.5, 131.4, 131.3, 123.1, 123.0, 122.8, 119.0, 115.5, 104.5, 96.4, 90.9, 90.2, 88.7, –0.10. Mass spectroscopy (MS) (electrospray ionization): m/z 392 $[M]^+$, 377 $[M - \text{CH}_3]^+$. Decomposition temperature $T_d = 281$ °C.

Dimethyl-4-[4-(4-trimethylsilylethynylphenylethynyl)phenylethynyl]phenylamine (2): Yield: 90 %. Anal. Calcd. for $\text{C}_{29}\text{H}_{27}\text{NSi}$: C, 83.40; H, 6.52; N, 3.35. Found: C, 82.82; H, 6.64; N, 3.36. ^1H NMR (300 MHz, CDCl_3): δ 7.50–7.37 (m, 10H), 7.65 (d, $J = 8.9$ Hz, 2H), 3.00 (s, 6H), 0.26 (s, 9H). ^{13}C NMR (75 MHz, CDCl_3): δ 150.3, 132.8, 131.9, 131.5, 131.4, 124.4, 123.4, 123.0, 121.8, 111.8, 109.7, 104.6,

96.3, 93.0, 91.3, 90.5, 87.2, 40.2, 29.7, -0.09. Fast-ion bombardment MS: m/z 417 $[M]^+$, 402 $[M-Me]^+$, 386 $[M-2Me]^+$, 372 $[M-NMe_2]^+$, 345 $[M-SiMe_3]^+$, 300 $[M-NMe_2-SiMe_3]^+$. $T_d = 245^\circ\text{C}$.

Dimethyl-[4-(4-[(triisopropylsilyl)ethynyl]phenylethynyl)phenylethynyl]phenylamine (3): Yield: 89%. Anal. Calcd. for $C_{35}H_{39}NSi$: C, 83.78; H, 7.83; N, 2.79. Found: C, 83.24; H, 7.61; N, 2.34. ^1H NMR (400 MHz, CDCl_3): δ 7.48–7.38 (m, 10H), 6.68–6.63 (m, 2H), 3.00 (s, 6H), 1.13 (s, 21H). ^{13}C NMR (100 MHz, CDCl_3): δ 150.3, 132.8, 132.0, 131.5, 131.3, 131.2, 124.3, 123.4, 123.0, 121.8, 111.8, 109.7, 106.7, 93.0, 92.9, 91.2, 90.6, 87.3, 40.2, 18.7, 11.3, 0.0. FAB MS: m/z 501 $[M]^+$. $T_d = 315^\circ\text{C}$.

For the bottom-contact devices, layers of the oligo(arylacetylene)s with thicknesses of ca. 50 nm were deposited on the Au-patterned SiO_2/Si substrates at room temperature under high-vacuum condition (3×10^{-4} Pa). For the top-contact devices, the active organic layer (50 nm) was deposited on 100 nm SiO_2 substrates followed by the deposition of a gold electrode on the top using different masks for organic and metal layers. The devices fabricated on untreated substrates were tested in a microprobe station to measure the current–voltage characteristics of the devices under ambient conditions.

Received: December 7, 2004

Final version: February 12, 2005

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A Lead-Free High-Curie-Point Ferroelectric Ceramic, $\text{CaBi}_2\text{Nb}_2\text{O}_9$ **

By Haixue Yan, Hongtao Zhang, Rick Uvic, Michael J. Reece,* Jing Liu, Zhijian Shen, and Zhen Zhang

High-temperature sensing technology is of major importance for chemical and material processing, as well as the automotive, aerospace, and power-generating industries. Electro-mechanical transducing materials are required to sense strains, vibrations, and noise under severe thermal conditions. Among the different types of acoustic and strain sensors, piezoelectrics offer the best candidates when one considers sensitivity, cost, and design.^[1] When an operating temperature of 400°C or greater is required, the choice of materials for high-temperature piezoelectric transducers is limited. Although some single crystals such as LiNbO_3 ,^[1] $\text{La}_3\text{Ga}_5\text{SiO}_{14}$,^[2,3] $\text{Sr}_2\text{Nb}_2\text{O}_7$,^[4] and $\text{A}_2\text{Ti}_2\text{O}_7$ (where A is La or Nd)^[5] are good commercial or potential candidates for high-temperature piezoelectric applications, the cost of single crystals is very much higher than that of polycrystalline piezoceramics.^[1,2] Another important ad-

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[**] This research was funded by QinetiQ Ltd under contract number CU004-16541 and the Swedish Research Council through grant 621-2002-4299.