Unique Role of Self-Assembled Monolayers in Carbon Nanomaterial-Based Field-Effect Transistors

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Molecular self-assembly is a promising technology for creating reliable functional films in optoelectronic devices with full control of thickness and even spatial resolution. In particular, rationally designed self-assembled monolayers (SAMs) play an important role in modifying the electrode/semiconductor and semiconductor/ dielectric interfaces in field-effect transistors. Carbon nanomaterials, especially single-walled carbon nanotubes and graphene, have attracted intense interest in recent years due to their remarkable physicochemical properties. The combination of the advantages of both SAMs and carbon nanomaterials has been opening up a thriving research field. In this Review article, the unique role of SAMs acting as either active or auxiliary layers in carbon nanomaterials-based field-effect transistors is highlighted for tuning the substrate effect, controlling the carrier type and density in the conducting channel, and even installing new functionalities. The combination of molecular self-assembly and molecular engineering with materials fabrication could incorporate diverse molecular functionalities into electrical nanocircuits, thus speeding the development of nanometer/molecular electronics in the future.

1. Introduction

Molecular-based optoelectronic devices have attracted broad attention over the past two decades because they exhibit many inherent advantages, such as mechanical flexibility, versatile chemical design and synthesis, low cost, light weight and solution processability. In particular, the thin film transistor (TFT) is one of the most important fundamental building blocks for electronic circuits. In general, in a TFT, carrier transport in the channel between source and drain electrodes occurs in at most the first few layers of the semiconductors, close to the dielectric layer adjacent to the gate electrode.^[1,2] This implies that a closely-packed defect-free 2D crystalline semiconducting monolayer should exhibit a bulk-like carrier mobility and current modulation.^[3-5] Meanwhile, in a field-effect sensor where the response is dominated by the electrostatic interaction between analytes and the conducting channel, the current modulation is limited by the semiconductor thickness. As a result, scaling the dimensions of semiconductors down to the monolayer will greatly improve the device sensitivity, making them highly suitable for sensing applications.^[6,7] Therefore, much effort has recently been made to scale down the thickness of the TFT devices as demonstrated by several groups.^[3,7-20] These studies utilized sophisticated molecular assemblies that spontaneously happen when the substrates are dipped into solutions of the active components to form an active part of TFTs, either the gate dielectric^[9,11,13–17] or the semiconductor layer.^[3-5,7,10,12] In the former case, TFTs built on self-assembled monolayers (SAMs) as the gate dielectric can increase the capacitive coupling through the gate insulators and function at a very low-operating voltage (as small as 2 V), thus reducing the power dissipation. In the latter case, if densely-packed SAMs are achieved, the performance of SAMTFTs is comparable to that of bulk materials.^[3–5] Because these devices are formed from a monolayer, their electrical properties significantly change when they are exposed to external stimuli, forming the basis for new types of environment and molecular sensing.^[7,21,22]

Carbon nanomaterials, especially single-walled carbon nanotubes (SWNTs) and very recently graphene, have also attracted much attention because of their exceptional physical, chemical and electronic properties. Graphene is a 2D crystalline monolayer made of sp^2 -hybridized carbon atoms arranged in a honeycomb lattice.^[23–25] By folding up a graphene sheet into a cylinder along a certain lattice vector, a well-ordered, hollow graphitic nanomaterial, classified as a SWNT, is formed.^[26] Both of these two allotropes of carbon have the simplest chemical composition and atomic bonding configuration in a 2D manner that maximizes their surfaceto-volume ratio. Each carbon atom on the nanocarbon surface is exposed to the environment and any small changes in the environment can result in drastic changes in the physicochemical properties of the nanocarbon device.^[7,21,22,27] In conjunction with their high conductivity, high chemical stability, and easy availability through bottom-up approaches, the environmental ultrasensitivity places carbon nanomaterials-based field-effect transistors (FETs) as a promising platform for creating new types of sensors and optoelectronic devices.^[28-31]



In this Review article, we describe a unique research in the incorporation of SAMs into carbon nanomaterials-based FETs. We begin with the formation of SAMs from different functional molecules. We then detail an efficient strategy of how to improve the TFT performances, applying SAMs as semiconducting layers and carbon nanomaterials as electrodes, respectively. Finally, we demonstrate the important role of SAMs as auxiliary layers in the modulation of the physicochemical properties of SWNTs and graphenes. However, this research field is a diverse and rapidly growing one. Having limited space and references, we will only be able to cover some of the major contributions with the most general applicability and highlight some important aspects, which were neglected in most previous reviews. Fortunately, there are a number of excellent previous review papers in the literature covering various aspects of carbon nanomaterials-based field-effect transistors, which can amend these deficiencies.[28,32-41]

2. Formation of SAMs

There are several articles reviewing the approaches of self-assembling functional monolayers on different substrates.^[42–46] In this review, we will only focus on the ways of forming densely-packed high-coverage SAMs used for improving the TFT performance and/or tailoring the properties of carbon nanomaterials, specifically SWNTs and graphene.

2.1. Covalent Binding Method

In carbon nanomaterials-based FETs, the surfaces that can be chosen for covalently forming SAMs are primarily limited to two: the electrode surface and the dielectric surface. For example, Shin's group has pre-modified the electrodes with SAMs through Au-S bonds leaving a positively- or negatively-charged end-group. Then the reduced graphene oxide (rGO) were selectively adsorbed on the gold electrodes by electrostatic attraction to form rGO field-effect transistors.^[47] Derycke's group has demonstrated the use of octadecanethiol

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SAMs on gold as a local gate for a single-walled carbon nanotube field-effect transistor (SWNT-FETs).^[48] In a FET, the surface of the dielectric layer plays an important role in SAM applications. The most frequently-used dielectric layer is silicon dioxide (SiO_2) with free hydroxyl groups on the surface, which can easily react with organosilanes, phosphonic acids, or acvl chlorides. Organosilanes, such as hexamethyldisilazane (HMDS),^[49] octadecyltrichlorosilane (ODTS),^[50] octadecyltrimethoxysilane (OTMS),^[51] octyltrichlorosilane (OTS),^[53] et al., are often used to create a dense hydrophobic buffer layer for passivating the surface and correspondingly minimizing the substrate effect.^[52-55] Guo and his coworkers demonstrated the surface reaction of acyl chlorides of hexabenzocoronenes (HBCs) with the activated SiO₂ surface to produce stable densely-packed semiconducting SAMs through an ester linkage.^[7] In some cases, to decrease the dielectric thickness, inorganic dielectric materials with larger permittivity (for example TiO_x , AlO_x and HfO_x) were used. Similarly, to passivate the hydroxyl groups on their surfaces, alkylphosphonic acids were usually used to form high-quality SAMs.^[56] It should be mentioned that the substrates need to be pre-activated in order to provide suitable reactive sites for the subsequent surface reactions. The most commonly-used activating methods are oxygen plasma or UV/ozone treatments to create active hydroxyl groups on the surfaces of inorganic dielectric layers.^[46]

2.2. Spin-Coating Approach

Molecules with an anchoring group can covalently bond to the surface to form the monolayers. However, it is not an essential requirement. It was found that symmetric molecules with long alkyl groups can form high-quality monolayer films through a simple spin-coating approach. For example, Mourran's group used α, ω -octyldodecylseptithiophene to form a monolaver on SiO₂ substrate at a critical concentration and relatively fast rotating rate.^[5] Nuckolls' and our group have also used liquid crystalline tetra(dodecyloxy)hexabenzocoronenes (HBCs) to assemble a columnar monolayer into the nanogaps between SWNT electrodes.^[7,21] Molecules suitable for this strategy in general have long alkyl groups symmetrically substituted at both ends. These long alkyl groups provide high solubility in common organic solvents, surface hydrophobicity and water resistance, and liquid crystallinity for molecules, of crucial importance to realize low-cost highperformance monolayer transistors for future switching and sensing applications.^[57-59] However, due to the weak interaction with the substrate, these monolayers are fragile when exposed to organic solvents.

2.3. Langmuir-Blodgett Technique

The Langmuir-Blodget (L-B) technique is usually used to prepare large-area ordered ultrathin films with well-defined architectures. Molecules suitable for the L-B technique are thought to be amphiphilic.^[60] However, typical amphiphilic molecules, usually a carboxylic acid with an alkyl group in the



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opposite site, will lead to carrier trapping by carboxylate.^[61] Copper phthalocyanine (CuPc) is not a typical amphiphilic molecule, but it has been demonstrated that CuPc dissolved in a mixture of trifluoroacetic acid and dichloromethane could form a stable monolayer at the air-water interface and could be deposited on substrates with Pc macrocycles oriented nearly perpendicular to the substrates.^[62–64] Recently, our group took advantage of this method to achieve highperformance CuPc monolayer transistors with high photoresponsivity, using either 1D SWNT or 2D graphene as electrodes.^[22,27] Therefore, the L-B technique proves to be an efficient method to form homogenous high-coverage denselypacked monolayers.

3. SAMs Acting as Active Layers, Carbon Nanomaterials Acting as Electrodes

In organic field-effect transistors (OFETs), with size reduction, the device performance is mainly hampered by the contact resistances with high injection barriers between the metal electrodes and the organic semiconductors (OSCs). Most commonly, gold is chosen as the charge-injecting metal because of its chemical stability and its work function that matches the energy level of OSCs in most cases, thus lowering the Schottky barriers.^[65] However, it is not suitable to the case when SAM is applied as the semiconductor layer. One main reason is that the SAM layer is commonly below 5 nm, the use of adhesion layer (commonly 5–10 nm) would inhibit charge injection and collection between gold electrodes and Table 1. Summaries of the roles/types of SAMs used in carbon nanomaterials-based $\ensuremath{\mathsf{FETs}}$

Role of SAMs	Function	Molecular type ^{a)}	Reference
Active Layer	Chemoresponsive behavior	HBCs	[7]
	Photoresponsive behavior	HBCs	[21]
		CuPc	[22,27]
Auxiliary Layer In SWNT-FETs	s SAM-based gate dielectric	POTS	[93]
		ODE@gold ^b)	[48]
		ODPA@AlO _x	[56]
	In-situ separation of m- & s-CNTs	APTES/PTEOS	[96,103]
		CUTS	[108]
In G-FETs	Controlling sub- strate effect	HMDS	[49]
		OTMS	[51,55]
		ODTS	[50]
		PBTS	[52]
		HMDS/OTS/ODTS	[53]
	Controlling the carrier type	PFTS/BTES/APTES	[136–138,142
		ZPP@rGO/Pt	[143]
	Forming graphene p-n junctions	APTES	[151]
		APTES/PFES	[152]

^{a)}The molecular type refers to the precursors used for forming SAMs; ^{b)}Without additional specification, molecules were assembled on the SiO₂ surface; HBC: hexabenzocoronene; CuPc: copper phthalocyanine; POTS: 18-phenoxyoctadecyltrichlorosilane; ODE: octadecanethiol; ODPA: *n*-octadecylphosphonic acid; APTES: 3-aminopropyltriethoxysilane; PTEOS: triethoxyphenylsilane; CUTS: 11- cyanoundecyltrimethoxysilane; HMDS: hexamethyldisilazane; OTMS: octadecyltrimethoxysilane; ODTS: octadecyltrichlorosilane; PBTS: 4-phenylbutyltrichlorosilane; OTS: octyltrichlorosilane; PFTS: perfluorooctyltriethoxysilane; BTES: butyltriethoxysilane; rGO: reduced graphene oxide; ZPP: Zinc(II) porphyrin.

SAMs. Several methods were proposed to solve the problem. de Leeuw's group used 5-10 nm titanium films as the adhesion layer. Then the electrodes were defined by standard photolithography and wet etching. The titanium layer at the edge of the contact was dissolved by HNO₃/HF etchant. The gold electrode was under-etched by about halt a micrometer. Then the collapsed gold electrode was contacted with the SiO₂. Hence, despite the use of the Ti adhesion layer, the charge injection occurred directly through the Au contact.^[3,4] Beyond the wet-etching of metal electrodes, another strategy to minimize the contact resistance was to replace the metal electrodes with several alternative materials, including conductive polymers,^[66] carbon nanotubes (CNTs),^[67-69] carbon nanotube/polymer nanocomposites,^[70,71] and graphene/graphene multilayers.^[22,72,73] In this section, we will focus our discussion on the two most commonly-used carbon nanomaterials, single-walled carbon nanotubes and graphene, as potential substitutes for gold source/drain (S/D) electrodes (Table 1).

3.1. SWNTs as Electrodes

It is well known that SWNTs are molecular chemicals which have the similar structures with organic molecules and are entirely composed of carbon atoms, a natural match to OSCs. In addition to this, SWNTs hold the suitable work function that matches the energy level of OSCs. Therefore, these remarkable properties implies that SWNTs can form the perfect contact interface between them and OSCs through π - π stacking, lower the Schottky barriers, and thus potentially work as efficient electrodes for making high-performance OFETs. The use of SWNTs as electrodes was pioneered by Qi and Tsukagoshi et al.^[67,74] They applied a high current through the channel to disconnect each metallic singlewalled or multi-walled carbon nanotubes that bridged the two metal leads. Then organic materials (pentacene or P3HT) were deposited to bridge the gap through vapor or solution processing. Both studies formed ultra-small OFETs with nanoscale channel lengths, proving that carbon nanotubes can be novel electrodes for molecular devices. However, it is hard to control the gap size by electrical breakdown in mass production. To overcome the problem, we developed a system to form SWNT electrodes by cutting them using precise local oxidation through a polymethylmethacrylate (PMMA) mask defined by e-beam lithography (**Figure 1**a–c).^[75] This process leaves two ends that are separated by a gap of ≤ 10 nm. One significant feature of SWNTs is that they are quasi-1D ballistic conductors that are intrinsically the same size as SAMs, thus offering the opportunity of detecting the subtle response of SAMs to external stimuli. Guo and his coworkers used SWNT point contacts detailed above as the source and drain electrodes to measure the current across a SAM between two ends of a cut SWNT.^[7] Contorted HBCs were used here because it is surrounded by alkyl groups, making it easy to self-organize into molecular stacks with exceptional semiconducting properties.^[8,12,76] Transistors made from HBC SAMs have shown high gate efficiency and large ON/OFF ratios (Figure 1d,e). In addition to these, their electrical properties changed significantly when exposed to electron-deficient molecules such as tetracyanoquinodimethane (TCNO). However, we failed to observe the photoresponsive behaviors in these monolayer transistors. To achieve the photoresponses, we further incorporated liquid crystalline HBCs into SWNT electrodes to make efficacious columnar FETs.^[21] The molecules used in this work are able to self-organize into columnar nanostructures with a diameter similar to that of SWNTs, thus allowing the investigation of the intrinsic properties of individual self-assembled columns. Interestingly, when the devices were exposed to visible light, photocurrents with an on/off ratio of >3 orders of magnitude were observed. In general, it is difficult to detect the photoresponsive properties of monolayers because of instantaneous detrimental charge recombination under light irradiation and the quenching of the photoexcited states of monolayer molecules exposed to the environments. However, in the current case, SWNT electrodes can form excellent interfaces contact with molecules and exhibit barrier-free-like injection. Therefore, the application of relatively small voltages yields efficient charge



Figure 1. a) Oxygen plasma introduced through a defined PMMA window affords precise cutting of SWNTs. b) Opening of a SWNT yields point contacts separated by ~2 nm. c) AFM image of a SWNT gap. Inset: height profile of an isolated tube. Reproduced with permission.^[75] Copyright 2006, AAAS. d) Monolayer model with three layers color-coded to show relative thickness. e) Monolayers of self-assembled stacks being probed with SWNT electrodes. Reproduced with permission.^[7] Copyright 2006, NAS.

injection in the gap area. This could significantly avoid detrimental charge recombination under light irradiation and the quenching effect by the environments, thus affording efficient charge transport through SWNT contacts at low voltages.

Both aforementioned works are the stark examples showing the possibility of installing new functionalities into electrical nanocircuits through the combination of molecular engineering and chemical self-assembly with materials fabrication, thus opening up a unique way for building new types of ultrasensitive optoelectronic devices for environmental and chemical sensing. For instance, a growing research interest in the field of optoelectronic devices is the development of solution-processed sensors with high sensitivity for low-cost, large-area sensing, high reproducibility, easy and environmental-friendly process. However, the detector sensitivities are in general limited by inefficient carrier dissociation and transport of materials and high contact Schottky barriers. To solve these problems, most recently, we detailed an approach, by which a new class of ultrasensitive solution (water)-processed photodetectors in the visible region based on high-performance photoresponsive OFETs at the molecular level were developed.^[27] These efficacious nanophotodetectors were formed from monolayers of a typical OSC, copper phthalocyanine (CuPc), using 1D SWNTs as point contacts (Figure 2a). Uniform CuPc monolayers were vertically transferred from the water/air interface onto the substrate surface through the conventional L-B technique. By applying the S/D bias voltage to metal contacts attached to the nanotubes and the gate bias voltage to the doped silicon as global back-gate electrode, we can tune the carrier density in the devices in combination with photoexcitation. Typically, the resultant nanodevices behaved as p-type, hole-transporting semiconductors (Figure 2b). Remarkably, we are able to measure their DC photoconductivity at room temperature in ambient atmosphere under light illumination (Figure 2c). We found that the device photoresponsivity was bias-dependent (Figure 2d). Varying S/D and gate biases results in the gradual improvements of the responsivity values, indicating the fine-tunability of the device photoresponsivity. Operating at low biases, the monolayer photodetectors exhibit responsivities greater than 10^8 A/W, detectivities greater than 7×10^{15} Jones, and high reproducibility. These results suggest that the three-terminal transistor geometry might be superior to the commonly-used two-terminal device structures for improving the photodetector performance.

3.2. Graphene as the Electrode

Graphene, a newly isolated form of carbon, holds a set of remarkable electronic and physical properties that make it ideal for use in nanoelectronics.^[23,25,28,77] In comparison with 1D SWNTs, graphene is a 2D, zero-gap semiconductor without diameter and chirality dependences. In combination with the intrinsic properties such as low resistivity, high chemical stability, high mechanical strength, and easy availability in very high crystallographic quality, these distinct and complementary advantages with SWNTs strongly suggest that graphene should be another type of electrode candidates for making a wide variety of organic optoelectronic devices.

The first example is that Liu's group deposited graphene on patterned thin films of Cu or Ag to serve as hybrid electrodes for pentacene FETs (**Figure 3**a).^[72] The presence of graphene decreases the work function of metal electrodes (and therefore the contact resistance between the electrodes and semiconductors), thus leading to a high carrier mobility of $0.53 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. This value is about ten times higher than that with the pure Cu or Ag contact electrodes. Recently, our group developed a method to directly use cut pristine metallic, single-layer graphene sheets as 2D nanogapped S/D electrodes through ultra-fine electron beam lithography and



Figure 2. a) Schematic illustration of ultrasensitive water-processed monolayer photodetectors with SWNTs as point contacts. b) Output characteristics of a typical p-type device. c) The time dependence of I_D as the lights with different power were switched on and off. Inset is the power dependence of the changes in I_D . d) Responsivities versus wavelength and calculated detectivities at $\lambda = 620$ nm and $\lambda = 702$ nm at various biases for a device. Reproduced with permission.^[27] Copyright 2011, RSC.

precise oxygen plasma etching (Figure 3b).^[78] After deposition of organic thin films (either P3HT or pentacene) as the semiconductors, these nanotransistors based on graphene

electrodes showed high-performance FET behaviors with bulk-like carrier mobilities, high on/off current ratio, and high reproducibility (Figure 3c).^[79] Furthermore, due to



Figure 3. a) Schematic illustration of the fabrication of OFETs with patterned graphene electrodes. Reproduced with permission.^[72] Copyright 2008. b) Device structure of P3HT transistors based on nanogapped graphenes as electrodes. c) Device characteristics of a representative P3HT thin film transistor based on 2D graphene contacts. $V_{\rm G} = 100$ to -5 V in -21 V steps. Reproduced with permission.^[78] Copyright 2009.



Figure 4. a) Schematic illustration of CuPC monolayer transistors based on nanogapped graphenes as planar contacts. b) Device characteristics of a representative CuPC monolayer transistor. c) Time dependence of I_D as the lights with different power were switched on and off. Inset is the power dependence of the changes in I_D . $V_D = -8$ V and $V_G = 0$ V. Reproduced with permission.^[22] Copyright 2010.

the presence of photosensitive organic materials, reversible and significant changes in current were observed when the devices were exposed to visible light. It should be mentioned that mass production of graphene from chemical reduction of graphene oxide (rGO) provides an inexpensive source for large-scale applications. Several groups have reported good performance of rGO electrodes used in solar cells^[80,81] and OFETs.^[73,82]

The L-B technique offers a promising and reliable method to prepare large-area ordered SAMs. For example, we and others have demonstrated the successful applications of the L-B technique to CuPc and conjugated polymers in producing ultrathin film OFETs.^[62-64] However, the charge carrier mobilities in these devices were low, namely about 10^{-7} ~ 10^{-3} cm²V⁻¹s⁻¹. This might be ascribed to the high contact resistance when gold was used as S/D electrodes and to the defects in the micrometer-long channels. In our further work, we employed single-layer graphene as S/D nanoelectrodes with channel gap of around 50 nm to overcome these difficulties (Figure 4a).^[22] The method gave almost 100 percent yields of the working monolayer transistors. At the high negative gate voltage bias, we observed the superlinear increase in the drain current $I_{\rm D}$ with S/D voltages $V_{\rm D}$ at low values; when $V_{\rm D}$ reaches -15 V, the current almost reaches saturation (Figure 4b). The calculated average carrier mobility is high (~0.04 cm²V⁻¹s⁻¹, one to three orders of magnitude higher than those based on metal electrodes) with high on/off ratio $(10^5 \text{ to } 10^6, \text{ three orders of magnitude})$ higher than those based on metal electrodes), and reliable reproducibility. Similar to the cases of SWNTs, we observed the reversible and stable photocurrents in our devices at room temperature in ambient atmosphere (Figure 4c). This is surprising because the photocurrent in the devices occurs within a single 1.3 nm-thick layer. These results prove that grapheme sheets as planar contacts can form excellent contact interfaces with molecules and exhibit barrier-free-like injection. Therefore, the application of relatively small voltages yields efficient charge injection in the gap area in our devices, avoiding detrimental charge recombination under light irradiation and the quenching effect by the environments. The concept developed here of incorporating molecular functionalities into molecular electronic devices by combining bottom-up self-assembly and top-down device fabrication should speed the development of nanometer/ molecular electronics in the future.

4. SAMs as Auxiliary Layers, Carbon Nanomaterials as Active Layers

In OFETs, SAMs with a wide range of anchoring groups (silanes, phosphonic acids, carboxylic acids, etc.) have been employed to modify the semiconductor/dielectric interfaces.^[42,45,83] This can improve the device performance by one or more of the following mechanisms: (1) passivating trap sites at the dielectric surface (e.g., -OH groups and dielectric ionic polarization),^[9,34,40] (2) improving the ordering and/or charge accumulation in the OSC through chemical SAM/ OSC interactions,^[33,34,37–39,84] and (3) matching the surface energy between the dielectric surface and the OSC.^[28,35,41] In this section, we will mainly overview the use of SAMs at the semiconductor/dielectric interfaces for controlling the properties of single-walled carbon nanotubes (SWNT-FETs) and graphene-based FETs (G-FETs) (Table 1).

4.1. SWNT-FETs

4.1.1. SAM-based Gate Dielectric in SWNT-FETs

Over the past decade, SWNTs have emerged as highly promising components of nanoscale electrical devices. Among them, SWNT-FETs have been thoroughly studied since 1998 due to their 1D character, nanometer scale, and exceptional performances.^[85-87] Most commonly, a highly doped silicon substrate covered with thermally grown SiO₂ serves as a macroscopic back gate. In order to achieve increased capacitive coupling, alternative gate dielectrics of ultrathin thickness^[88,89] or high dielectric permittivity^[90,91] have been investigated while the corresponding FETs often suffer from pronounced hysteresis in the transfer characteristics.^[92,93] Recently, organic-based gate dielectrics, in particular organic-inorganic bilayers^[94] and self-assembled multilayer gate dielectric grafted onto SiO₂,^[95] have been studied for transistors composed of nanotube networks. Devices reported in these work showed a greatly reduced hysteresis, but inferior on/off ratio and mobility. In addition, such devices display relatively large gate leakage currents. To further improve the device performance, the use of SAMs as gate dielectric might be a good choice as evidenced in the field of OFETs.^[13,14,17] Weitz's group pioneered the study in SWNT-FETs by using a 2 nm-thick silane-based SAM grafted onto SiO₂ surface (Figure 5a).^[93] Their devices



Figure 5. a) AFM image of the SWCNT-FET device structure with a 2 nm-thick silane-based SAM grafted onto SiO₂ surface. Reproduced with permission.^[93] Copyright 2007, ACS. b) Schematic device representation of a double back-gate SWNT-FET. Reproduced with permission.^[48] Copyright 2008, AIP.

displayed remarkable performances and reduced hysteresis in the transfer characteristics. In particular, they operated with a gate-source voltage of only -1 V and exhibitd good saturation, large transconductance, and small hysteresis (≤100 mV), as well as a very low subthreshold swing (60 mV/dec) under ambient conditions. The SAM-based gate dielectric opens the possibility of fabricating transistors operating at low voltages and constitutes a major step toward nanotube-based flexible electronics. Then, Derycke's group studied the operating mechanism and performances of a double back-gate SWNT-FET using a SAM of octadecanethiol grafted onto a local gold electrode as organic dielectric (Figure 5b).^[48] The use of an additional global back gate and the control of the environment (air or vacuum) allow tuning the polarity of the device from p-type to n-type. P-type devices display high $I_{\rm on}/I_{\rm off}$ ratios, drastically reduced hysteresis, very steep subthreshold

slope, and band-to-band (BTB) tunneling, whereas variations of the local gate potential has no influence on the drain-source current of n-type devices. This latter result is explained by the critical impact of the SAM dipole moment which drastically modifies the local-gate work function.

As shown above, using SAMmodified gate dielectric is an efficient way to improve the SWNT-FET performances, such as low-voltage, large transconductance, steep subthreshold swing, and large on/off ratio. However, their integration into electrical circuits with large signal gain and good stability still needs to

be demonstrated. Weitz's group demonstrated that these important goals can be achieved with the help of a bottomgate device structure that combines patterned metal gates (20 nm thick layer of aluminum) with a thin gate dielectric (3.6 nm thick layer of aluminum oxide) based on a molecular self-assembled monolayer (2.1 nm thick of *n*-octadecylphosphonic acid) (**Figure 6**).^[56] The devices show excellent operational and shelf life stability performance without degradation during 10 000 switching cycles and during storage under ambient conditions for more than 300 days. The device structure also allows the implementation of unipolar logic circuits with good switching characteristics.

4.1.2. In-situ Separation of m-CNTs and s-CNTs

To find full use in electronics, SWNTs need to be efficiently separated by electronic types (metallic carbon nanotubes



Figure 6. False colored AFM image of a completed carbon nanotube FET. The gate stack is sketched in the top inset. A zoom showing the channel of the FET in greater detail is shown in the bottom inset. Reproduced with permission.^[56] Copyright 2009, ACS.

(m-CNTs) and semiconducting carbon nanotubes (s-CNTs)). However, almost all of the currently available technologies can only produce SWNTs in a mixture of both m- and s-types during synthesis. This coexistence in as-produced samples dramatically decreases the device performance; therefore, SWNTs should be separated before they are integrated into devices. Two main separation or purification processes have been developed during the past decade to solve this problem.^[96] The first one is selective destruction by, for example, nitronium ion attack,^[97] gas-phase etching reactions,^[98] weak oxidative carbon sources,^[99] and ultraviolet irradiation.^[100] The second one is solution-based methods like dielectrophoresis,^[101] density-gradient-inducing centrifugation,^[102] selective adsorption of chemicals,^[103] and agarose-gel-based separation.^[104] Both techniques can separate SWNTs effectively, but the former introduces damages to SWNTs, while the latter can only work for very short SWNTs and both suffer from difficulty of scaling up.

The pioneering work by Papadimitrakopoulos has shown selective absorption of amines towards SWNTs.^[105] The following theoretical and experimental studies also demonstrated

that aromatic molecules like the phenyl-terminated silanes prefer to interact and bind selectively to m-SWNTs.[106,107] Though the mechanism remains unclear, this strategy has already been exploited for separation. For example, Zhang's group developed a simple method to separate s-SWNTs from m-SWNTs by using SAM-modified polydimethylsiloxane (PDMS) thin films such as 'scotch tape' (Figure 7a).^[96] Interestingly, A-scotch tapes which were treated with 3-aminopropyltriethoxysilane (APTES) could remove s-SWNTs leaving m-SWNTs on the substrate, while P-scotch tapes which were treated with triethoxyphenylsilane (PTEOS) could remove m-SWNTs leaving s-SWNTs on the substrate. This can be explained by the first-principle calculation results which suggest that the amine-terminated surfaces have stronger interaction with s-SWNTs (with binding energies of 203 and 214 meV in m-SWNTs and s-SWNTs, respectively), while phenyl-terminated surfaces have stronger interaction with m-SWNTs (with binding energies of 432 and 416 meV in m-SWNTs and s-SWNTs, respectively).

Recently, Bao's group successfully translated the selectivity to interactions at substrate surfaces.^[103]



Figure 7. a) Schematic of SWNT separation using P- and A-scotch tapes to selectively remove m- and s-SWNTs respectively, and leave their counterparts on the sapphire substrates. Reproduced with permission.^[96] Copyright 2011. b) The dielectric (300-nm SiO₂ on a heavily doped Si gate) is functionalized by either an amine-terminated (surface A) or phenyl-terminated (surface B) silane. c) The SWNT solution is subsequently dispensed onto the spinning self-assembled monolayer–modified substrate and dried, followed by source (S) and drain (D) gold electrode deposition. Shown here is the top-contact device structure. Upon spincoating, AFM tapping-mode topography images (10 mm by 10 mm, z scale = 10 nm) of the nanotubes applied under identical conditions on amine (top) and phenyl (bottom) surfaces reveals that density and alignment are a direct function of surface chemistry. Reproduced with permission.^[103] Copyright 2008, AAAS.

The SiO₂ dielectric surface was functionalized by either an amineterminated or phenyl-terminated silanes to form wellordered SAMs (Figure 7b). Then the SWNT solution was subsequently dispensed onto the spinning SAM-modified substrate and dried followed by S/D gold electrode deposition. The spin-assisted alignment and density of SWNTs are tuned by different surfaces that effectively vary the degree of interaction with SAM-modified surface functionalities in the channel. This leads to a self-sorted SWNT network in which nanotube chirality separation and simultaneous control of density and alignment occur in one step during device fabrication (Figure 7c). Further work demonstrated that the amine, and specifically the lone pair, is crucial in controlling any level of sorting in solution-deposited nanotube networks.^[69] However, amine silanes are extremely sensitive to atmospheric and preparation conditions leading to the difficulty in producing complete monolayers consistently and reproducibly. As a candidate, functional nitrile groups have been demonstrated to serve as a satisfactory replacement for amine.^[108] Not only is the SAM formation more predictable and reproducible, but SWNTs interact very strongly with nitrile groups. The amine group can donate electron density and charge to SWNTs, effectively dedoping them and reducing their performance while the nitrile group, with reduced electron donation ability, is thought to adsorb to the SWNTs strongly without reducing the air-induced p-type doping providing greater densities, higher current carrying capacity, and lower threshold voltages.

4.2. Graphene-Based FETs

Many scientists in the electron-device community are currently active in graphene research because of its possible applications in many emerging areas such as graphene-based electronic devices.^[28,109] In fact, graphene has been considered as a candidate material for post-silicon electronics according to the strategic planning report by the International Technology Roadmap for Semiconductors (ITRS).^[110] However, tuning the properties and the bandgap of graphene in a controlled and practical fashion is the primary challenge the scientific community is facing at this time. Detailed below are the main strategies developed for controlling the substrate effects, improving the device performance, and building functional graphene-based FETs, using SAMs as a unique tool.

4.2.1. Controlling the Substrate Effects

It is well-known that graphene-based FETs fabricated on dielectric substrates, commonly SiO₂/Si substrates, typically exhibit a field-effect mobility being multiple orders lower than the intrinsic graphene mobility.^[111,112] Moreover, transport properties of such graphene devices suffer from hysteretic behaviors under ambient conditions, as well as asymmetries between electron and hole transport.^[113–118] Though systematic studies are still lacking and are also difficult to carry out, evidences show that these limitations are caused by so-called substrate effects.^[54] for instance, charged impurity scattering,^[116] extrinsic scattering by surface phonons,^[111]

corrugation or doping by residual adsorbates.[113,120] As a result, high quality substrates to accommodate graphene devices have been suggested to be beneficial,^[111] vet finding such materials remains a challenge. The groups of Hong and Dean have used epitaxial ferroelectric dielectric $(Pb(Zr_{0.2}Ti_{0.8})O_3 (PZT))^{[121]}$ and hexagonal boron nitride (h-BN)^[122] as novel substrates for graphene devices. The charge carrier fabricated on these substrates are found to be 70 000 and 64 000 cm² V⁻¹ s⁻¹, respectively. However, the existing commercially available silicon dioxide (SiO₂) substrates are compatible with the conventional Si-based electronics^[123] and are also widely used in graphene research because of the convenient and mature production process, low leakage current, small surface roughness, best optical contrast, and low cost.^[55,124] Therefore, it is more useful to utilize surface/interface engineering to modify these substrates.

Smet's group pioneered the work by using a hydrophobic SAM of hexamethyldisilazane (HMDS) between graphene and the SiO₂ substrate (Figure 8a).^[49] The HMDS was thought to serve as a producible template that prevents the adsorption of dipolar substances providing a chemically well-defined and hydrophobic surface. Mobility on the modified substrates was increased two-fold, and the scattering effects were still to be elucidated. Low intrinsic doping levels and suppressed hysteretic behavior were also obtained even in ambient air. In another work from the Xu's group, by using a SiO₂/Si substrate modified by SAMs of OTMS, they have obtained high-quality graphene devices with a low intrinsic doping level.^[51,55] The high mobility could reach 47 000 cm² V⁻¹ s⁻¹. This could be attributed to the monolavers consisting of closely packed and long inert alkyl chains, which effectively minimized the densities of unintentionaldopants and charge traps on the substrate surface. As a result, both charged impurities and remote interfacial phononscattering could be reduced. With a ODTS SAM-functionalized SiO₂/Si substrates (Figure 8b). Chen's group obtained the electron and hole mobility as high as 60 000 cm² V⁻¹ s⁻¹.^[50] Most interestingly, the remarkable magnetotransport of graphene devices with pronounced quantum Hall effect (QHE) and strong Shubnikov-de Haas (ShdH) oscillations also confined the high quality of graphene on this ultrasmooth organic SAMmodified platform.

However, the aforementioned works are based on exfoliated graphene flakes, which are unsuitable for large-scale application. To this end, a chemical vapor depositition (CVD) process now enables a reliable way for the scalable and manufacturable production of graphene for large-area device integration. Recently, the Liu's group systematically studied the interface physics and interface engineering for large-scale CVD graphene transistors by applying the SAM of 4-phenvlbutvltrichlorosilane (PBTS) at the dielectric/graphene interface.^[52] Hysteresis of the CVD graphene-based FETs as measured at room temperature was found to be dominated by charge injection from graphene to the dielectric/graphene interface (Figure 9). For the gate bias stress stability of both graphene-based FETs with and without SAM interface engineering, the Dirac point shift was well described by the stretched exponential model. The fitting results clearly indicated a superior interface quality with less trapping effects



Figure 8. a) Left: Schematic of the contacted graphene sample on top of an HMDS self-assembled layer.; Middle: Three-dimensional structure of the HMDS molecule; Right: Optical image of the contacted graphene flake. Reproduced with permission.^[49] Copyright 2010, ACS. b) Left: Schematic representation of graphene on an ODTS-modified SiO₂/Si substrate; Right: QHE of graphene on ODTS-modified substrates at B = 9 T and T = 2 K. Reproduced with permission.^[50] Copyright 2012, ACS.

involved with the phenyl-SAM engineered graphene devices. Another systematic study of the effect of SAMs on the performance of graphene-based FETs was conducted using CVD-grown large-area graphene by the Cho's group.^[53] The graphene/SiO₂ interfaces were treated with hydrophobic SAMs of three organoalkylsilanes with varying alkyl chain lengths (C1, C8 and C18, respectively). As the SAM alkylchain length increased, substrate-induced doping of graphene decreased, and thus the devices exhibited higher hole/electron mobilities with lower Dirac point voltages. Changes in the electrical performances in the presence of SAMs could be explained by charged impurity scattering, indicating that charged impurities in graphene could be tuned by the alkyl chain length of a SAM inserted between the graphene and the SiO₂ substrate. They also demonstrated that charged impurity scattering arises from the chemical and physical properties of the substrate surface as well as from the intrinsic quality of graphene. Therefore, appropriate surface chemistries at graphene/dielectric interfaces are essential for enhancing the electrical properties of graphene-based FETs.

4.2.2. Controlling Carriers in Graphene

Pristine single-layer graphene exhibits a standard ambipolar behavior with a zero neutrality point in a FET device on standard SiO_2 substrates, and the ambipolar properties limit its electronics applications. In this regard, it is necessary to control the charge carrier type and density of graphene without destroying its characteristic electronic structure. Two main strategies have been made to modify the electronic structure of graphene to make n- and p-type FETs.^[113,125–133] The first one is direct substitution with boron and nitrogen in the graphene lattice to create p- and n-type doping.^[128,130,132,133] However, heteroatom substitutions break the symmetrical structure of the graphene lattice and lead to a 10 to100-fold decrease in graphene's carrier mobilities. The second method includes physically or chemically doping graphene with small molecules.^[116,125,126,129] Physically adsorbed molecules are not stable and are easily desorbed under vacuum or heat, while chemical functionalization suppresses the mobility of graphene due to the newly formed sp^3 C-C bonds.

To overcome these drawbacks, SAMs are chosen as an unique approach to control the carrier type and density of graphene without destroying its characteristic electronic structure. This is because that surface modification with SAMs to tune the charge carrier accumulation at the dielectric/semiconductor interfaces have been thoroughly studied in OFETs.^[84,134,135] Several groups have employed different types of SAMs of perfluoroalkylsilane, alkylsilane, aminoalkylsilane, and aminoarylsilane, formed on SiO₂ substrates to control the carrier type and density of graphene (Figure 10a,b).^[136-138] For single layer graphene on perfluoroalkylsilane-SAM, the stiffening of the Raman G-band indicates a large downshift of the Fermi level (~-0.8 eV) by accumulated hole carriers. Meanwhile, aminoarylsilane-SAM accumulated electron carriers, which compensate the hole carriers doped by adsorbed molecules under the ambient atmosphere in graphene (Figure 10c,d).^[136] As a consequence, a wide range of Dirac point shift have been obtained, from -18 V (H₂N-SAMs, n-doping) to +30 V (F-SAMs, p-doping).^[137] It has been demonstrated that the dipole alignment of the SAM molecules can produce a built-in electric field and thereby modify the carrier density of OFETs.^[84,134,139] Similarly, Yokota's group ascribed the sharp contrast of doping behavior between the graphene/(NH₂C₃H₆-SAM, F₁₃-SAM) and the graphene/NH₂C₆H₄-SAM to the difference in dipole moment of SAMs as consequence of inductive effect of N and F atoms and the resonance effect of = NH_2^+ group (Figure 10c,d).^[136] However, since the amine group bears a



Figure 9. a) A general bottom-gated graphene FET structure with phenyl-SAM interface engineering. b) Illustration of the physical mechanisms for the hysteresis behavior of graphene-based FETs at different gate biases. Reproduced with permission.^[52] Copyright 2010, ACS.

lone pair of electrons and the ammonium group is positively charged, H₂N-SAMs and H₃N⁺-SAMs induced doping in graphene transistors cannot be understood simply by using the built-in electric field model. A charge transfer mechanism, arising from the position of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of the molecule with respect to the Dirac-point of graphene and orbital hybridization, was proposed to explain small molecule-induced doping. The amine group in H₂N-SAMs can donate its lone pair to graphene's channel, increasing electron carrier density and inducing n-doping. In H₃N⁺-SAMs, the ammonium cations are positively charged, which is anticipated to withdraw electrons from the graphene channel, decreasing electron carrier density and inducing hole doping. This explanation is consistent with the discussions from the Dai's group, which suggested that the possible charge transfer between carbon nanotubes and aminopropyltriethoxysilane or polyethylene imine molecules could happen and thus result in the efficient n-doping in carbon nanotubes by the electron-donating ability of the amine groups.[140,141]

The Wang's group investigated charge transfer between graphene and the substrate by measuring the surface potential of graphene flakes using KPFM, which illustrated the distribution of carriers in different graphene layers as well as the formation of dipoles at the interface.^[142] The Fermi level of single layer graphene on the modified substrates could be tuned in a range from -130 to 90 meV with respect to the Dirac point, corresponding to the doped carrier concentrations up to 10¹² cm⁻². Besides dielectric surface modification, the modification of the S/D electrode interfaces with SAMs is also possible. Recently, Rao's group reported a unipolar operation in rGO-based FETs via modification of the S/D electrodes interfaces with SAM of zinc(II) porphyrin molecules.^[143] The dipolar zinc(II) porphyrin molecules at the rGO/platinum S/D interface results in an increase of the electron injection barrier and a reduction of the hole-injection barrier. The electron and hole injection barriers were calculated to be 2.2 and 0.11 eV, respectively, indicating a higher barrier for electrons, compared to that of holes. A reduced gate modulation in the electron accumulation regime in rGO devices with SAM shows that unipolar rGO-based FETs can



Figure 10. a) Chemical structures of four silane coupling agents used to fabricate SAMs. b) Schematic illustrations for the preparation of graphene/SAMs. c) The shift of the Fermi energy of graphene and the graphene/SAMs vs the dipole moment (μ) of the corresponding SAM molecules. The experimental results of $\Delta E_{\rm F}$ and the theoretical results of $\Delta E_{\rm F,calc}$ are indicated as circles and squares, respectively. The insets are the optimized structures of the theoretically calculated models for graphene and the graphene/SAMs. The arrows show the dipole moments of them. d) An illustration for the physical picture of the carrier doping mechanism in graphene/SAMs. Reproduced with permission.^[136] Copyright 2011, ACS.

be attained using a low-cost, solution-processable fabrication technique.

4.2.3. Forming Graphene p-n Junctions

Graphene p–n junctions have already displayed new and exciting phenomena such as Klein tunneling, where electrons

traveling perpendicular to the junction experience zero resistance,^[144] fractional quantum Hall transport,^[145] and Veselago lensing, where diverging electron waves are refocused by the junction.^[146] Therefore, great efforts have been made to fabricate graphene p-n junctions including multiple electrostatic gates,^[145,147] chemical treatment by gas exposure,^[117] polymer-induced doping,[113,148,149] and electronic modification of the substrates.^[150] In the previous section, we have demonstrated the use of SAMs on SiO₂ substrates to control the charge accumulation at the dielectric/graphene interface, resulting p-doping or n-doping of graphene. Is it possible to utilize two different kinds of SAMs to controllably dope graphene and form p-n junctions in graphene? This has been demonstrated by the work from the Graham's group (Figure 11a).^[151,152] This method used SAMs that covalently bond to the substrate rather than graphene, resulting in thermally stable doping without altering its structure.^[152] Monolithic and patterned 3-aminopropyltriethoxysilane (APTES) layers are used to create n-doped graphene, while perfluorooctyltriethoxysilane (PFES) were used to create p-doped graphene (Figure 11b). Simultaneous use of these groups in a FET device results in formation of two separate Dirac points, as indicative of a graphene p-n junction. This method minimizes doping-induced defects and results in thermally stable graphene p-n junctions.

5. Conclusion

In this article, we highlighted the unique role of SAMs in carbon nanomaterials-based FETs. SAMs, formed by either the covalent or noncovalent methods, can function as active layers for building high-performance, stimuli-responsive monolayer transistors using SWNTs or graphene as electrodes. This is because SWNTs and graphene can form the perfect contact interfaces with SAMs and thus lower the contact resistance. On the other hand, the incorporation of functional SAMs into the dielectric/semiconductor interfaces can tune the substrate effects for improving the FET performance and/or selectively separating SWNTs with different types, control the carrier type and density in the conducting channel, and even install new functionalities in carbon nanomaterials-based FETs. The strategies demonstrated in this review should provide new insights into designing novel SAM-based device structures for studying the intrinsic mechanism of interface phenomena to elucidate the interplay among parameters that control charge transfer, the doping effect, and the property of optoelectronic devices.

Although SWNT and graphene share some common properties, such as high conductivity and high chemical stability, each has its own advantages and disadvantages. For example, SWNTs are 1D and can be produced in bulk quantities while large-scale production of high-quality graphene is still difficult. In contrast, owing to its 2D nature without diameter and chirality dependences graphene is ideal for forming planar electrodes for OFETs and graphene could be produced using traditional lithographic techniques down to the nanoscale. These distinct and complementary capabilities of SWNT or graphene FETs could open up new opportunities for the



Figure 11. a) Schematic of the graphene p–n junction formed by SAM-induced asymmetric doping. b) Drain-source current versus gate voltage for graphene/SiO₂ (black), graphene/APTES/SiO₂ (green) and graphene/PFES/SiO₂ (red) after being heated at 200 °C for 3 h under nitrogen environment. Reproduced with permission.^[152] Copyright 2012, ACS.

development of new types of optoelectronic devices. We expect that the combination of molecular self-assembly and molecular engineering with materials fabrication could incorporate diverse molecular functionalities into SAMs-based carbon FETs, thus speeding the development of nanometer/ molecular electronics for a wide variety of applications ranging from future practical logic circuits and optoelectronic devices to ultrasensitive chemo/biosensors and medical diagnosis.

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