



Poly(9-vinylcarbazole)–graphene oxide composite field-effect transistors with enhanced mobility

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ABSTRACT

We report on a solution processed hybrid organic field-effect transistors (OFETs) incorporating both organic semiconductor – poly(9-vinylcarbazole) (PVK) and graphene oxide (GO) flakes. We have found that PVK:GO hybrid OFETs with Au–Al source–drain electrodes exhibit the mobility as high as 3.8–6.7 cm² V⁻¹ s⁻¹ under concentration of the GO flakes varies from 3 up to 9.5 wt.% respectively. The on/off ratio of PVK:GO hybrid OFETs is found to be ~10²–10³ at source–drain and gate operating voltages below ~5 V. The reversible switching of the source–drain current (from low to high *I*_{SD} and back) is found at positive and negative *V*_{SD} ~ 12–13 V in the PVK:GO OFET structures (GO ~ 3 wt.%) with Au–Al source–drain electrodes. The possible mechanism of enhancing field-effect mobility in the PVK:GO OFET structures is discussed.

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1. Introduction

Solution-processed organic semiconductors offer great potential for low-cost, high-scale manufacturing of flexible electronics based on organic field-effect transistors (OFETs) such as flexible organic light-emitting diodes display drivers, radio frequency identification tags, memory chips and chemical sensors [1]. Organic and polymeric semiconductors used in OFETs typically exhibit rather high on/off ratios, but demonstrate low charge carrier mobility as compared with inorganic materials that limit the range of possible applications. Development of new solution-processed organic and polymeric semiconductors and materials on their basis with high charge carrier mobility remains an important goal for organic electronics. Unlike organic semiconductors carbon-based materials such as

graphene or carbon nanotubes exhibit very high mobility but usually have suffered from relatively low on/off ratio in the OFET structures on their basis [2,3]. In this connection graphene-based polymer nanocomposites are one of the most promising and recent technological developments that combine unique properties of graphene and polymer materials in one nanohybrid material [4]. These nanohybrid materials show considerable improvement in electronic and optical properties that cannot be achieved using conventional composites or pure polymers [5]. In addition the use of hybrid (semiconducting polymer/inorganic nanoparticles) materials as active layers of OFETs also leads to remarkable mobility increase (up to ~2 cm²/V s) and threshold voltage reduction with increase of the concentration of inorganic nanoparticles [6,7]. In order to enhance the effective mobility in OFETs the hybrid thin-films containing organic semiconductors (PQT-12 or P3HT) and graphene flakes have been demonstrated recently [8]. Graphene is a two-dimensional lattice of carbon with single atom thickness and thus it is

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very attractive due to its outstanding electronic, mechanical and physical properties [2]. It was shown that few-layer and single-layer graphene flakes can provide good connection between the crystalline regions of the semiconductor film. This allows increasing an effective mobility up to $\sim 0.6 \text{ cm}^2/\text{V s}$ in PQT-12/graphene FETs [8]. The on/off ratio appears to decrease as the mobility begins to increase in such hybrid OFETs. This behavior can be explained by effect of percolation of low-dimensional graphene flakes at low density. When there are enough graphene mixed into the organic semiconductor film in order to enhance mobility, the density of the graphene flakes appears to approach the percolation threshold. This results in limiting the maximum mobility values achievable before the percolation threshold of graphene flakes and reduces the on/off ratio of such a hybrid (semiconducting polymer/graphene flakes) OFETs. Moreover graphene flakes dispersed in soluble organic semiconductor aspire to agglomeration that may reduce the charge carrier mobility. One may suggest that the conduction channel of an OFET covered by the semiconducting polymer/graphene oxide (GO) flakes composite might affect the mobility of the polymer/GO-based OFET due to, for instance, templating of the polymer molecules by the GO flakes. The charge carrier mobility of OFETs based on the polymer/GO flakes composite films would be expected to show higher values than those without addition of GO. From this point of view nanocomposites based on poly(9-vinylcarbazole) (PVK) mixed with graphene oxide flakes (PVK:GO) are rather promising, since they have different ways of polymerization, fabrication, and dispersion [9–11]. Moreover PVK:GO composite films with relatively low GO concentration (GO ~ 1 –3 wt.%) demonstrate conductivity switching behavior in sandwich configuration [12]. It is of interest to investigate electrical and optical properties of such PVK:GO composite films with low GO concentration as active layers of OFETs especially since the performance of pure PVK OFETs (a charge carrier mobility $\sim 4.8 \cdot 10^{-9} \text{ cm}^2/\text{V s}$) and pure GO FETs have been considered by other authors previously [13–15].

In this paper, we present the developing of hybrid OFETs consisting of both organic semiconductor – poly(9-vinylcarbazole) and GO flakes (concentration of GO ~ 0 –9.5 wt.%). It was shown that PVK:GO hybrid FETs with Au–Al source–drain electrodes exhibit the mobility as high as 3.8 – $6.7 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ under the concentration of the GO flakes increases from 3 up to 9.5 wt.% respectively. The on/off ratio of PVK:GO hybrid FETs is about $\sim 10^2$ – 10^3 at source–drain and gate operating voltages below $\sim 5 \text{ V}$. The reversible switching of the source–drain current is found at both: positive and negative $V_{SD} \sim 12$ – 13 V in the hybrid PVK:GO OFETs with GO $\sim 3 \text{ wt.}$ %. It was assumed that incorporating of GO into semiconducting polymer matrix is a promising route to enhance the performance of OFETs with relatively low operating voltages, probably, due to templating of the PVK molecules by the GO flakes.

2. Materials and methods

In this work poly(9-vinylcarbazole) (PVK), molecular weight $M_w \sim 1.1 \cdot 10^6$, band gap $E_g \sim 3.6 \text{ eV}$, was chosen

as the organic semiconductor because of its good air stability. PVK polymer used in our study was purchased from Sigma–Aldrich and used without further purification. As a second component of the composite film the GO flakes with average dimensions range of about $\sim 500 \times 500 \text{ nm}$ (plus–minus 100 nm) and thickness 1.5–2.5 nm have been chosen. GO was purchased from AkKoLab Company (Moscow) and used as received. Fig. 1a and b provides the chemical structure of PVK and structure of a single layer of GO (proposed in Ref. [16]) used in our experiments. We have fabricated the PVK:GO hybrid FETs which comprise a n-Si/SiO₂/Au/PVK:GO/Al structures. A heavily doped n+ silicon substrate was used with 200 nm thermally grown SiO₂ as gate dielectric with thermally evaporated gold (Au) and aluminum (Al) electrodes on top. The distances between the Au–Al and Au–Au electrodes were $\sim 7 \mu\text{m}$ and $\sim 15 \mu\text{m}$ respectively and the width of the electrodes was about $\sim 1 \text{ mm}$. PVK polymer and GO flakes were dissolved and dispersed in chloroform respectively, and then they were mixed together and subjected to ultrasonic treatment for 10 min by using ultrasonic homogenizer (Bandelin Sonopuls HD 2070, $f \sim 20 \text{ kHz}$). The mixtures with different concentration ratios of components – from 0 to 9.5 wt.% of GO – were drop-cast or spin-coated (at 3000 rpm) onto a Si/SiO₂ substrate with Au and Al electrodes. The composite films were dried and heated at 70–80 °C in N₂ atmosphere for 15 min. The thickness of the drop-cast composite films was about $\sim 0.6 \mu\text{m}$ according to Atomic Force Microscopy (AFM) results. The atomic composition of the films was tested by means of X-ray photoelectron spectroscopy (XPS) measurements using the Escalab 250xi instrument (Thermo Fisher Scientific Inc.). The base pressure during the XPS experiments was kept below 10^{-7} Pa . The samples for XPS measurements: pure PVK, pure GO and PVK:GO (GO $\sim 3 \text{ wt.}$ %) have been deposited to Si–SiO₂ substrates at 300 K without heating. Similar PVK:GO film have been deposited to the Si–SiO₂ substrate with subsequent heating up to 80 °C for 15 min (similar to OFET preparation procedure). The films thickness for XPS experiments was about $\sim 2 \mu\text{m}$. Absorption spectra were studied using Cary-50 (Varian) spectrometer. The samples for absorption measurements were drop cast onto quartz substrates (the film thickness $\sim 1 \mu\text{m}$). Detailed microstructure of these films was studied by AFM using P47-Solver NT-MDT. A LGI-21 pulse laser ($l = 337.1 \text{ nm}$, $E_i > 10^{-4} \text{ J}/\text{cm}^2$, $t \sim 10^{-8} \text{ s}$) was used to excite the photoluminescence (PL). The laser beam was focused onto the sample using the quartz lens. The PL spectra at 300 K were analyzed using a SPM-2 spectrometer (spectral resolution $\sim 2 \text{ nm}$) and a photomultiplier with a spectral sensitivity range 300–850 nm. The PL light collection was about $\sim 100\%$. The current–voltage (I – V) characteristics of hybrid PVK:GO-based OFETs were measured in vacuum ($3 \cdot 10^{-3} \text{ torr}$) in the dark at 300 K using a holder of liquid N₂ cryostat and a dc electronic computer controlled measuring system with a Keithley 6487 picoammeter/voltage source and AKIP-1124 programmable voltage source. The source–drain and gate voltages were varied between -15 V and $+15 \text{ V}$ in variable steps. The effective field-effect charge carrier mobility, μ_{FET} , of hybrid PVK:GO-based OFET was extracted from the I – V s in the saturation and weak

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