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Effects of Gold Nanoparticles on Pentacene Organic Field-effect Transistors

Keanchuan Lee, Wei Ou-Yang, Martin Weis*, Dai Taguchi, Takaaki Manaka, Mitsumasa Iwamoto

Department of Physical Electronics, Tokyo Institute of Technology
2-12-1 O-okayama, Meguro-ku, Tokyo 152-8552, Japan
Phone: +81-3-5734-2191 E-mail: iwamoto@ pe.titech.ac.jp

1. Introduction

Nanoparticles (NPs) hold many novel physical and chemical properties owing to the fact that surface atoms have a very high surface area to volume ratio and have possible future applications in electronics, optics, memory devices and so on. The small dimensions of NPs lead to small capacitance, which can be roughly calculated with a model of the concentric spherical capacitor using material parameters, $4\pi\epsilon_o\epsilon_r a$, where a is radius of sphere. The double-layer capacitance of metallic NPs encapsulated in organic insulator matrix is so small (attofarad) that the single electron charging to and from NPs occurs at room temperature. This quantized charging has been reported for the voltammetry measurement of gold NPs dissolved in the electrolyte solution and diffusing to the electrode [1], and in films of NPs attached to chemically modified electrodes [2] The charge storage in NPs attracted attention of applied research to develop NP-based nonvolatile memory. Incorporation of NPs to the organic device had been reported [1-2] and the charge storage was also described by the charge trapping phenomenon [3]. Preparation of OFET by incorporating TiO_2 NPs into PVP as gate composite which acts as high-k gate insulator had been reported by Chen et al[4-5]. Here we are using gold NPs and blended into PVA with different NPs concentrations as gate composite in OFET. We might expect the increase of channel conductivity in pentacene as well as effective dielectric constant of gate insulator in a manner same as the case of TiO_2 NPs embedded in PVP. Beyond that electrostatic charging of Au NPs in PVA might directly has effect on carrier injection, in a way similar to the case of electrostatic dipolar layer embedded on the surface of gate insulator [6]. Keeping in mind these, effects of gold NPs on pentacene OFET were studied and attention was paid to the carrier transport in steady-state (Current-Voltage Measurement) based on estimated contact resistances, effective mobilities, threshold voltage and so on.

2. Experiment

Gold NPs (20 nm) with concentration of 1mg/cm^3 in water was employed in this experiment (British Biocell). To prepare the gate insulator with different *wt*% of gold NPs the concentration of PVA was being adjusted. The main reason of using PVA as gate insulator in this experiment is not only because of its good insulating properties but also of its water solubility that makes it a good candidate. Since the concentration of gold NPs is fixed, to prepare the gate insulator solutions with different *wt*% of gold NPs the PVA *wt*% in the gold NPs solution must be varied. However, higher concentration of gold NPs will result in lower *wt*% of PVA. Therefore, to get identical thickness of PVA layer in different samples for comparison, the spinning and exhaust speed as well as the time must be adjusted. The samples' thicknesses were then measured by Dektak profilometer. The blended solution showed no aggregation of NPs with uniform red color. It was then spin-coated on 100 nm SiO_2 on highly doped Si substrate with gold as bottom electrode. The film was pre-annealed at 90 ^oC for 30 minutes to evaporate the solvent (water in this case) and finally annealed at 120 ^oC for 1 hour. The annealing was performed under N_2 environment. Pentacene with thickness of 100 nm was then thermally evaporated as organic semiconductor layer under shadow mask with a rate of 0.5 Å/s and finally gold was evaporated through the shadow mask and used as source and drain electrodes (patterned top-contact OFET). The channel length/width *L/W* were 40-100 μm / 3 mm. Fabricated pentacene OFET with gold NPs gate insulator is shown in Fig. 1. the current-voltage characteristics were measured by the Keithley Source Meter. Capacitance- and conductance-voltage measurement were also performed by Solartron Impedance Spectrometer. All measurements were carried out under ambient atmosphere.

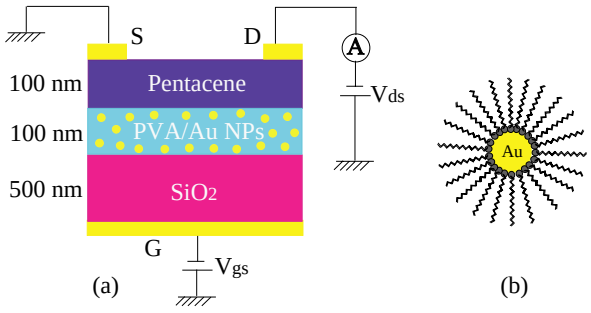

Figure 1 (a) Sketch of the top contact structure of organic field-effect transistor (OFET) with PVA/Au NPs gate insulator layer with *I-V* measurements and (b) the stabilized gold NP structure (GC20, British Biocell). The diameter is about 20 nm.

3. Results and discussion

Figure 2 illustrates the dependence of contact resistance with respect to biased gate voltage. We can observe that incorporation of NPs had decreased the contact resistance. This should be discussed based on the space charge effect contributing to decrease potential drop below the electrode. However we should know the reason behind the increased

*Institute of Physics, Slovak Academy of Sciences, Dúbravskácesta 9, 845 11 Bratislava 45, Slovak Republic

space charge field and one of the reason might be due to carrier trapping. The origin of the traps can be attributed to the gold NPs embedded in PVA, in the way same as composite system of Maxwell-Wagner effect element [7]. Note that it is known such system possess memory effect [8]. The trapped carriers might help to increase electric field below the source electrode and therefore the carrier injection is improved. The potential drop is represented as the electrical equivalent circuit of the OFET by the contact resistance element ($R_c=V_{drop}/I_{ds}$) [9,10], thus decay of potential drop represents decrease of the contact resistance.

Figure 2 (b) shows the dependence of mobility against the applied voltage at different NPs concentration. We can see that there are slightly increase of carrier mobility when NPs concentration increases but the increase is not significant (less than an order) and it lies within the experimental error. Main reason is that carrier path for injected holes is formed in pentacene close to the PVA/Au NPs layer.

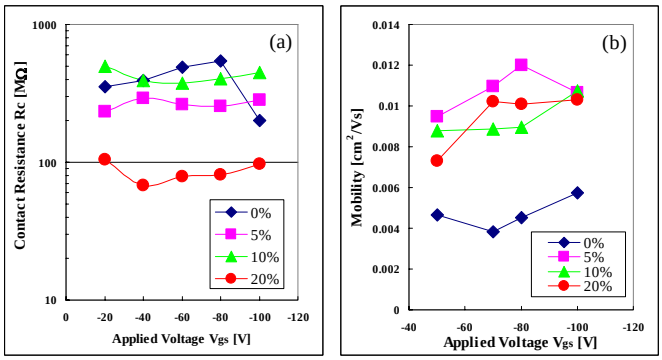

Figure 2 (a) Contact resistance (b) Mobility as a function of applied voltage for different NPs concentration

Table 1: Summary of device parameters for various Au NPs concentrations evaluated from transfer characteristics at drain-source voltage (V_{ds}) of -70 V and gate-source voltage (V_{gs}) sweep +20 to –70 V. The channel length is L = 60 μm.

Concentration	0%	5%	10%	20%
V_{th} (V)	25	3.1	0.15	-14.9
On/off ratio (10^4)	7.08	4.08	5.40	7.70
Mobility (10^{-3} cm^2/Vs)	3.11	9.54	8.62	9.18
R_c (MΩ)	500	260	380	80
Capacitance (nF/cm^2)	9	8.23	6.5	8.9
ΔQ (μC/cm^2)	0	0.18	0.16	0.36

Table 1 illustrates the summary of 60 μm samples with applied voltage of 70V. As NPs concentration increases, the threshold voltage shifted more to negative, which indicates the trapping of holes.

There are no decrease of on/off ratio and significant increase of mobility (at same order) which indicates the effects of conductivity can be omitted. In other words, the effect of gold NPs is more on trapping phenomenon instead of improving channel conductivity which can be further supported by increased trapped charged density, ΔQ, and decreased contact resistance, R_c, relative to pure PVA sample. There are no significant changes in gate insulator capacitance, which can be supported by Bruggeman mixing formula [11] where there will be only significant change of effective dielectric constant of a mixture at high concentration of inclusions even if the inclusions have very high dielectric constant. Finally we should note the electric field formed by the trapped carriers below the source electrode can compensate the applied electric field. Therefore further investigation is needed e.g. electric field induced SHG (EFISHG).

4. Conclusions

We had demonstrated the incorporation of gold NPs into gate insulator as composite served as trapping centers and by designing the concentration we can actually control the threshold voltage and trapped charge of the device. Gold NPs actually helped the carrier injection and improved charged accumulation thanks to its charging phenomenon. However there is not much influence on channel conductivity in pentacene as well as capacitance of gate insulator based on the same order of on/off ratios and effective mobilities when NPs concentration increases.

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