



Metal nanoparticles in organic field-effect transistor: Transition from charge trapping to conduction mechanism

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ABSTRACT

Self-assembled monolayers of metal nanoparticles (NPs) are envisioned for various devices and have been investigated for possible applications. However, organic envelope of NPs which is required for self-assembling must be often removed prior further device fabrication. Here, we report on effect of ozonolysis on monolayer of silver NPs (Ag NPs) with size of 8 nm and its impact on Ag NPs utilization in organic field-effect transistor. It was found that Ag NPs covered by organics serve like a traps and removal of insulating organics decreases number of traps and consequently increases Ag NPs monolayer conductivity.

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1. Introduction

Organic field-effect transistors (OFETs) are envisioned as logic devices in organic electronics applications [1] because of the low-cost fabrication. However, their applications can also be expanded to other fields such as electroluminescence (EL) or memory devices [2]. The performance of organic devices which depends on the charge transport is one of the most studied phenomena in device physics in last decade [3,4], and the variations in device geometries to improve the measured current especially in OFETs [5] have made the comparison of device parameters complicated. Nevertheless, the charge storage effect in the device, *i.e.* memory effect, had also gained attention of scientists and researchers.

On the other hand, nanoparticle (NP) is considered as a concentric spherical capacitor according to common model of double-layer charging [6]. Hence, one of the promising ways to store charges in the devices is by introducing metal NPs into the devices, and their charge storage properties had attracted attention in application-based research to develop NP-based non-volatile memory. The NPs which act as spherical nanocapacitors with high charging energy make the charge storage favorable [7]. Hence, the retention time of memory device stands for the charge relaxation time of the nanocapacitors. The small size of NPs leads to extremely low value of capacitance and observation of the single-electron charging to and from an NP at room temperature. Note that this charging can be interpreted as the Maxwell–Wagner

(MW) charging of the double-layer system [8], by considering that the charging was caused from the difference in carrier spreading time between NPs and surrounding organic materials. Incorporation of NPs into the organic device had been reported [9–11] and the charge storage was also described by the charge trapping phenomenon [12]. Many approaches had been done to obtain charge retention properties where NPs are added into the devices as nanocomposite gate insulator [2], nanocrystal [13], sandwiched between gate-insulator and blocking layer [10] as well as Langmuir–Blodgett layer [14]. It was found that air–liquid interface provides an ideal platform to produce self-assembling of NP films [15] especially by using the Langmuir technique [14]. The NP film was created on the air–liquid interface and subsequently deposited onto the solid substrate. Compared with blending of NPs into gate insulator which acts as a composite which had been reported mainly as high-*k* materials [16], the self-assembled monolayer (SAM) sandwiched between semiconductor-insulator interface provides us a simple and direct way to understand the interface phenomenon, because we can control the density of NPs more easily and also can gather information regarding the effects of trapped charges at the interface. However, the preparation of NP SAM requires presence of organic envelope (surfactant monolayer encapsulating metal NP) which suppresses the agglomeration of NPs into larger clusters and plays a vital role in the self-assembly process. This residual organic which represents unwanted parasitic resistance in applications is needed to be removed by non-invasive removal techniques like thermal annealing in vacuum or ozonolysis [17].

In this study we report the effect of the ozonolysis of the silver NPs (Ag NPs) on the performance of pentacene OFET with Ag NPs monolayer on the organic semiconductor–gate insulator interface. The influence on the organic envelope as well as metal NP core is recorded and the total film resistance is evaluated. The pentacene OFETs used here are used for

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an illustration of the NP influence on organic device properties. The experimental results are well supported by the Maxwell–Wagner model [18] analysis.

2. Experimental details

The Ag NPs (8 nm) encapsulated in oleic acid/oleylamine were synthesized according to the method published in detail elsewhere [19]. NPs were dissolved in chloroform (Sigma-Aldrich) prior the spreading on the water subphase (18.2 M Ω cm) of the Langmuir trough using a microsyringe (Hamilton, Reno, NV). The monolayer was allowed to equilibrate for 30 min before the compression. This time was found to be sufficient for the solvent evaporation and monolayer formation. Subsequently the Ag NPs monolayer was deposited on the solid substrate by the modified Langmuir–Schäffer (horizontal lifting) technique [20]. The details on NP size and spatial distribution were reported earlier [21]. The ozonolysis was done by the exposure to the ozone generated in oxygen rich atmosphere aided by UV irradiation (UV/ozone chamber, Nippon Laser & Electronics Lab, Japan) for a specific time.

Organic devices used in experiments were top-contact pentacene OFETs. Heavily-doped Si wafers with a 500 nm thick thermally prepared silicon dioxide (SiO₂) insulating layer were used as the base substrates. The Ag NPs monolayer was deposited onto Si wafers prior to the deposition of pentacene (100 nm in thickness). During the deposition of pentacene, the pressure was kept at less than 10⁻⁴ Pa and the deposition rate was fixed at 0.5 Å/s, monitored using quartz crystal microbalance. After the deposition of pentacene, gold electrodes (source and drain electrodes) with a thickness of 100 nm were deposited on the pentacene surface. The designed channel length (*L*) and width (*W*) were 30–100 μ m and 3 mm, respectively. Prepared

devices were characterized by the standard steady-state current–voltage measurement using Keithley 2400 SourceMeter. The Ag NPs monolayer conductance measurements were done on the Ag NPs deposited on the glass substrate with gold electrodes fabricated in the same manner as for the OFET device. The conductance measurement results shown here were recorded for 30 μ m long channel. All measurements were performed in laboratory ambient atmosphere.

For the optical extinction spectra or electrical conductivity measurements Ag NPs monolayers were deposited on glass substrates (Matsunami S-0313 Neo). Optical spectra were recorded by Jasco V-570 spectrophotometer in the double beam operation in the spectral region (400–800) nm at room temperature.

3. Results and discussion

Effect of ozonolysis on the Ag NPs organic envelope was evaluated by the change of the contact angle of the water as depicted in Fig. 1(a). The oleic acid is dissociated into fragments as azelaic acid and 9-oxononanoic acid [22,23]. The molecular structure of oleic acid as well as ozonolysis products is depicted in Fig. 1(b) (geometry optimized by the MOPAC software, parametric model 3 (PM3) Hamiltonian). Oleic acid dissociation to aerosols and fragments with higher polarity [24] leads to lower water contact angle. The ozonolysis also influences the NP metal core at the same time. The optical extinction spectra Ag NPs monolayer shown in Fig. 1(c) reveals presence of the surface plasmon peak at the wavelength of \sim 500 nm. However, the exposure to the ozone causes several effects like the oxidation of the silver NP core (Ag to AgO_x), change of effective dielectric constant of surrounding NP, as well as NP order in monolayer. Hence, the surface plasmon effect is

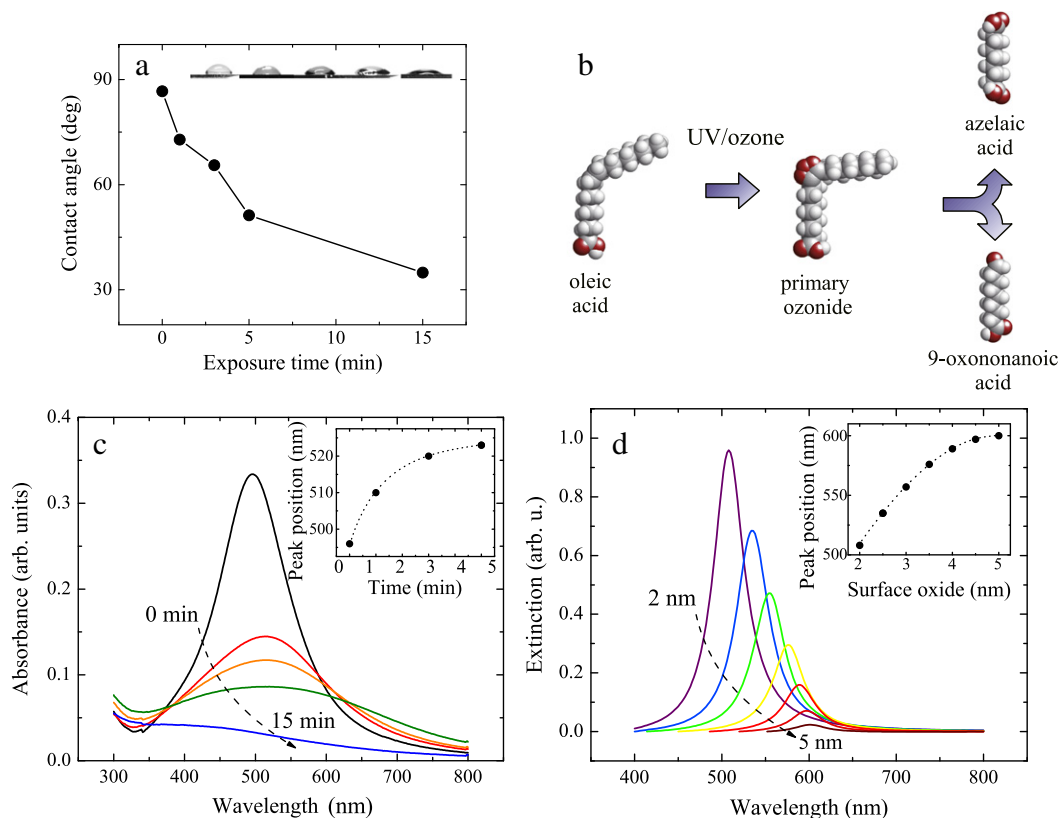


Fig. 1. (a) Effect of UV/ozone treatment time on the Ag NPs monolayer contact angle. (b) Optimized geometry of the oleic acid molecule and its fragments after the ozonolysis. (c) Extinction spectra of Ag NPs monolayer as a function of UV/ozone treatment time. (d) Extinction spectra of Ag NP calculated in accordance with Mie's theory for of NPs with Ag core and Ag₂O shell for different surface oxide thickness. Insets in (c) and (d) depict shift of the extinction peak position. Broken lines are to guide the eyes only.

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