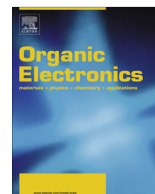




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## Organic Electronics

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# Improved thermal stability in photochromism-based optically controllable organic thin film transistor



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## ARTICLE INFO

## Article history:

Received 8 April 2014

Received in revised form 23 May 2014

Accepted 27 May 2014

Available online 7 June 2014

## Keywords:

Photochromism

Optical switching

Organic thin film transistor

## ABSTRACT

Thermally induced structural change in photoisomerization molecules is a serious obstacle to the development of optically controllable organic field-effect transistors (OFETs). This is because the thermal relaxation of molecular structures degrades photo-induced change in drain current and removes the memory function. To deal with this issue, a naphthopyran (NP) derivative, namely 3,13-dihydro-3-(4-triphenylaminy)-3,13-diphenylbenzopyrano[5,6-a]carbazole (NP-TPAC) was tested that displays pseudo p-type photochromism at room temperature. The NP-TPAC-doped poly(triarylamine) (PTAA) film exhibited a reversible change in transistor properties; the drain current was reduced by ultraviolet (UV) and returned to its original value by visible (VIS) light irradiation. Importantly, no change in the drain current was observed at room temperature for more than 30 h under dark conditions. This was because the open-ring *trans-trans* (TT) isomer of NP-TPAC is thermally stable owing to the CH- $\pi$  interaction and the steric force exerted by the phenyl ring of the carbazole unit onto the double bond responsible for the thermal back reaction. In other words, the thermal stability of photochromism-based optical devices can be greatly improved by adopting an appropriate molecular design.

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## 1. Introduction

Recently, organic field-effect transistors (OFETs) integrated with specific functionalities, e.g., light-emitting OFETs [1,2], pressure sensors [3,4], and gas sensors [5] have attracted considerable attention. A typical example of a functional OFET is an optically controllable OFET, which makes it possible to develop new optical devices

such as photo-controlled memory devices and optical sensors. For these purposes, photoisomerization molecules have been employed, in which the transistor properties were optically modulated in association with the photoisomerization [6–13]. Orgiu et al. have developed an optical switching transistor by doping photochromic diarylethene (DAE) molecules into poly(3-hexylthiophene), where the DAE molecules worked as hole trap sites [6]. The open-closed ring isomerization of DAE molecules involved a variation in the highest occupied molecular orbital (HOMO) level, which triggered the optical switching of the drain currents. Samori et al. have developed an optical switching transistor by using a self-assembled monolayer of photochromic azobenzene molecules, where the carrier injection barriers were tuned by varying the energy levels, which

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was accompanied by the *cis*–*trans* isomerization of azobenzene [7]. We previously achieved the optical control of transistor properties by doping spiropyran (SP) molecules dispersed into a PTAA channel layer [14]. In our case, an ionic polarized state of SP worked as a carrier scattering site, allowing effective suppression of the drain current. This mechanism based on ionic state scattering has a considerable advantage because it is applicable to all types of host semiconducting polymers regardless of the energy levels of the host/guest materials.

However, a significant problem remains as regards these photochromic molecule applications, namely the thermal instability of the photoisomerized state [15–17]. For example, the ionic polarized open-ring isomer of SP thermally relaxed to a non-polarized closed-ring isomer even at room temperature without any light irradiation to degrade the photo-memory property. In fact, we observed the thermal relaxation of the drain current in the SP/PTAA transistor [14]. The drain current suppressed by UV light irradiation returned to its original value within 10 h, despite the sample being kept at room temperature under dark conditions.

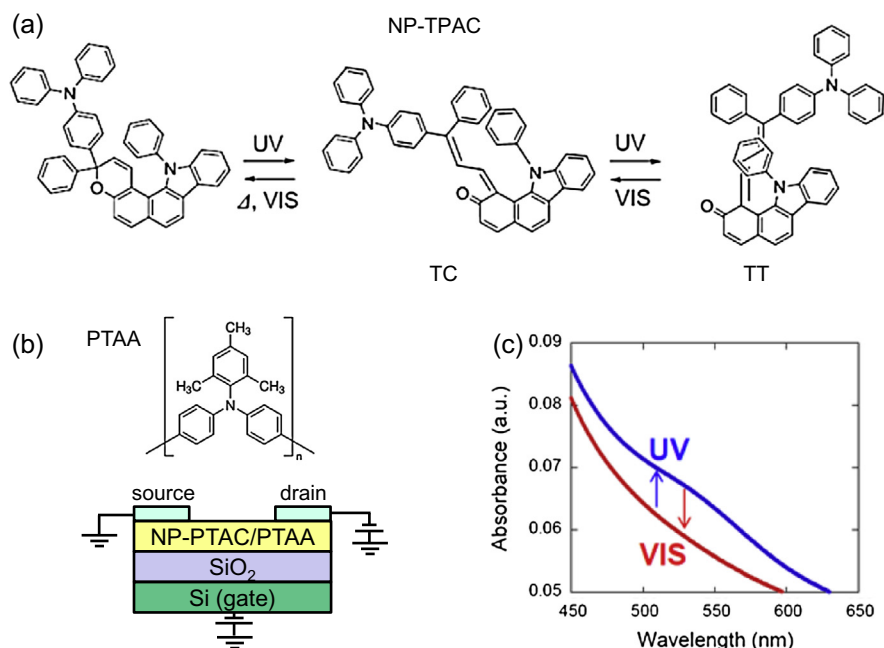
In this study, we used a recently synthesized naphthopyran (NP) derivative, namely 3,13-dihydro-3-(4-triphenylaminyl)-3,13-diphenylbenzopyrano[5,6-a] carbazole (NP-TPAC), which possesses a triphenylamine and carbazole moiety (Fig. 1(a)) [18]. The NP-TPAC-doped PTAA film worked as a transistor channel, which optically switched the drain current in the same way as our previous SP/PTAA transistor. An important point is that the thermal stability of the photo-memory property was greatly improved because of the high thermal stability of the

open-ring *trans*–*trans* (TT) isomer at room temperature. No change in the drain current was observed for more than 30 h under dark conditions. Our approach holds promise for the development of photochromism-based optical memory devices with long retention times.

## 2. Experimental procedure

The chemical structures and the photochromic reactions of NP-TPAC are shown in Fig. 1(a). The synthesis of this molecule is detailed in the Supporting Information. Upon UV irradiation, the closed-ring isomer undergoes a C–O bond cleavage to yield an open-ring *trans*–*cis* (TC) isomer, which in turn produces an open-ring *trans*–*trans* (TT) isomer via the photoisomerization of one double bond. The thermal half-life of the TT form was estimated to be 77 days in toluene solution.

Fig. 1(b) shows the configuration of a transistor and the chemical structure of PTAA. Highly doped p-type Si wafers with a 200-nm-thick SiO<sub>2</sub> layer were used as substrates, and their surfaces were terminated by hexamethyldisilazane to improve wettability and to reduce the number of charge trapping sites at the interface between the SiO<sub>2</sub> and NP-TPAC/PTAA layers. The Si substrate and SiO<sub>2</sub> layer work respectively as a gate electrode and a gate insulator layer. A 1:1 (weight ratio) mixture of PTAA and NP-TPAC dissolved in toluene at 1 wt% was spin-coated (1000 rpm for 1 min at room temperature) on the substrate. The samples were then annealed at 100 °C for 1 h to remove residual solvents. The thickness of the NP-TPAC/PTAA film was estimated to be 55 ± 10 nm with a surface profilometer



**Fig. 1.** (a) Chemical structures of NP-TPAC. Open-ring isomers of NP-TPAC are reversibly transformed into a closed-ring isomer and vice versa by alternating UV and VIS light irradiation. The *trans*–*trans* (TT) isomer is thermally stable at room temperature. (b) Schematic illustration of a photochromism-based transistor and the molecular structure of PTAA. (c) Reversible change in the absorption spectra of an NP-TPAC/PTAA thin film on a quartz substrate upon UV and VIS light irradiation.

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