



# Interface effect in pentacene field-effect transistors from high energy proton beam irradiation



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## ABSTRACT

We report the effect of irradiation using 10 MeV high energy proton beams on pentacene organic field-effect transistors (OFETs). The electrical characteristics of the pentacene OFETs were measured before and after proton beam irradiation with fluence (dose) conditions of  $10^{12}$ ,  $10^{13}$ , and  $10^{14}$   $\text{cm}^{-2}$ . After proton beam irradiation with fluences of  $10^{12}$  or  $10^{13}$   $\text{cm}^{-2}$ , the threshold voltage of the OFET devices shifted to the positive gate voltage direction with an increase in the current level and mobility. In contrast, for a high proton beam fluence condition of  $10^{14}$   $\text{cm}^{-2}$ , the threshold voltage shifted to the negative gate voltage direction with a decrease in the current level and mobility. It is evident from the electrical characteristics of the pentacene OFETs treated with a self-assembled monolayer that these experimental observations can be attributed to the trapped charges in the dielectric layer and pentacene/SiO<sub>2</sub> interface. Our study will enhance the understanding of the influence of high energy particles on organic field-effect transistors.

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## 1. Introduction

Organic electronic devices have been widely explored due to potential advantages including a variety of material choices, an easy fabrication process, low-cost mass production, flexibility, and printability [1–5]. Especially, organic field-effect transistors (OFETs) are important elements in contemporary electronics due to their wide range of potential applications such as identification tags, electronic bar codes, and active matrix elements for displays [6–9]. So, many research efforts have been devoted to the characterization and understanding of OFETs made from various organic materials, as well as the enhancement of the electrical performance of these devices [10–13]. The electrical characteristics of OFETs such as current levels, on/off ratio, mobility, and operational turn-on voltage (or threshold voltage) play critical roles in understanding the device operation and developing optimized devices, especially with respect to the charge injection and transport through the interface.

In particular, to fully tailor the fascinating electrical properties of OFETs into next generation electronics, we need to control the threshold voltage and mobility. The threshold voltage and mobility values of OFETs are highly affected by the semiconductor–dielectric interface [14–16]. In that context, there have been many studies about modification of the semiconductor–dielectric interface in various ways. For example, Kang et al. showed the enhanced device performance of rubrene OFETs by using graphene and hexagonal boron nitride as the electrodes and gate dielectric layers recently [17]. Due to charge-trap free clean hexagonal boron nitride and graphene interface, field effect mobility increased and hysteresis was suppressed. Furthermore, there have been studies on improving the electrical properties of OFETs through modifying inorganic surfaces or interfaces using functional molecules, including self-assembled monolayers (SAMs) [18–20]. These inserted SAMs could reduce the density of interface trap states and improve the morphology of the active pentacene layers. Others have modified the operation properties of OFETs by irradiation with ultraviolet (UV) light or gamma rays [21,22]. UV light irradiation resulted in electron trapping at the pentacene/dielectric interface and the pentacene OFET's electrical characteristics changed due to the slow release of trapped electrons. On the other hand, high energy gamma ray irradiation induced positive hole trapping in the

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SiO<sub>2</sub> layer of pentacene OFETs.

But, as far as we know, there have been no studies on high energy proton beam irradiation effects on the pentacene/SiO<sub>2</sub> interface of pentacene OFETs to control the threshold voltage or mobility. Considering several previous studies about tuning the electrical characteristics of pentacene OFETs by irradiating the high energy proton beams to inorganic FETs [23–25], we could expect tuning the electrical characteristics by proton beam irradiation. It is well-known that when high energy beams of charged particles are incident on FET devices, beams induce the trapped charges in the dielectric layer and the semiconductor/dielectric interface. And these trapped charges affect carriers of the active material of devices [26,27]. As a result of these trapped charges, the electrical properties of devices can be tailored. Besides, high energy proton beam irradiation experiments on pentacene OFETs could be a good opportunity to see the application of pentacene OFETs in aerospace radiation environment [28].

In this study, we investigated the electrical characteristics of pentacene OFET devices on SiO<sub>2</sub>/p++Si substrates through 10 MeV high energy proton beam irradiation. We systematically characterized the electrical properties of pentacene OFET devices before and after proton beam irradiation with different beam irradiation time conditions. We also studied the effect of proton beam irradiation on pentacene OFETs when the dielectric surface was coated with a passivating octadecyltrichlorosilane (OTS) SAM. The proton beam irradiation effects on the pentacene OFETs were analyzed based on the interplay between the proton beam irradiation-induced trapped charges inside the SiO<sub>2</sub> dielectric layer and at the pentacene/SiO<sub>2</sub> interface.

## 2. Experimental

### 2.1. Device fabrication

For the pentacene OFET device fabrication, a SiO<sub>2</sub> (270 nm-thick)/p++Si substrate was prepared and cleaned by dipping in an ultrasonic bath of acetone, isopropyl alcohol, and de-ionized water for 5 min at each step. The source and drain electrodes were prepared by depositing Au (30 nm)/Ti (5 nm) layers using an electron beam evaporator with a deposition rate of 0.5 Å/s at a pressure of  $\sim 10^{-7}$  Torr. The active channel was prepared by depositing a 60 nm-thick pentacene film using a thermal evaporator with a deposition rate of 0.5 Å/s at a pressure of  $\sim 10^{-6}$  Torr. For the molecular treatment on the pentacene OFETs, we deposited an octadecyltrichlorosilane (OTS) self-assembled monolayer (SAM) on the SiO<sub>2</sub> layer surface by immersing the sample in a silane solution (0.1 wt. %) in anhydrous toluene for  $\sim 12$  h under a N<sub>2</sub> atmosphere. The chemically treated samples were cleaned in toluene for 20 min and dried by blowing N<sub>2</sub> gas.

### 2.2. Proton beam irradiation

The proton beam irradiation experiments were performed using an MC-50 cyclotron at the Korea Institute of Radiological and Medical Science. The proton beam had an energy of 10 MeV, an average beam current of 10 nA, and a beam uniformity of approximately 90%.

### 2.3. Characterization of materials

X-Ray diffraction (XRD) measurements were taken using a D8-ADVANCE (Bruker) with Cu K $\alpha$  radiation at the Center for Materials Analysis at Seoul National University. AFM measurements were taken using an NX 10 AFM system (Park Systems).

### 2.4. Measurement of electrical characteristics

We measured the electrical characteristics of pentacene OFET devices using a semiconductor characterization system (Keithley 4200-SCS) and a probe station (JANIS ST-500) at room temperature in a vacuum ( $\sim 10^{-4}$  Torr).

## 3. Results and discussion

Fig. 1a shows a schematic illustration of proton beam irradiation on a pentacene OFET device. The fabrication process of the pentacene OFET devices is as follows. First, a Si wafer with a 270 nm-thick SiO<sub>2</sub> dielectric layer was cleaned by a standard solvent cleaning process. Au (30 nm)/Ti (5 nm) layers were then deposited as the source and drain electrodes on the Si wafer through a patterned shadow mask using an electron beam evaporator. Next, we deposited a 60 nm-thick pentacene active layer using a thermal evaporator. More details of the device fabrication process are explained in the Experimental Section and in the Supplementary Data (Fig. S1). Fig. 1b shows optical microscope images of the fabricated pentacene OFET devices. The right one is the image of entire device and the left one is enlarged one. The channel length and width of the OFETs are 100 and 300  $\mu$ m, respectively.

Following the electrical measurements of the pentacene OFET devices, a 10 MeV proton beam was irradiated onto the top-surface of pentacene OFET devices using a proton beam facility (MC-50 cyclotron) at the Korea Institute of Radiological and Medical Sciences (see the schematic image of Fig. 1a). Different proton beam irradiation times of 20, 200, and 2000 s were used, which correspond to a total fluence (or dose; the number of irradiated particles per unit area) values  $\Phi$  of  $\sim 10^{12}$ ,  $10^{13}$ , and  $10^{14}$  cm<sup>-2</sup>, respectively. The electrical characteristics of each device were systematically measured and compared before and after the proton beam irradiation. For statistical analysis, we measured more than five devices for each proton beam irradiation condition.

Fig. 1c–f present the representative electrical characteristics of the pentacene OFET devices before and after the devices were irradiated with proton beams. Fig. 1c shows the output characteristics (source-drain current versus source-drain voltage,  $I_{DS}-V_{DS}$ ) for a pentacene OFET measured at gate voltages ( $V_G$ ) varying from 30 to  $-30$  V with a step of 10 V before and after the proton beam irradiation with a fluence of  $10^{12}$  cm<sup>-2</sup>, corresponding to a beam irradiation time of 20 s. Fig. 1d shows the semilogarithmic plot of transfer characteristics (source-drain current versus gate voltage,  $I_{DS}-V_G$ ) measured for the same device at a fixed source-drain voltage ( $V_{DS}$ ) of  $-40$  V before and after proton irradiation with a fluence of  $10^{12}$  cm<sup>-2</sup>. This is also plotted on the linear y-axis in the inset of Fig. 1d. Fig. 1e and f show the output and transfer characteristics, respectively, measured for another pentacene OFET device before and after proton beam irradiation with a higher fluence condition of  $10^{14}$  cm<sup>-2</sup>, corresponding to a longer irradiation time of 2000 s. Notably, we observed that the pentacene OFET devices under different proton beam irradiation conditions exhibited distinct electrical behaviors. When the devices were irradiated with a fluence of  $10^{12}$  cm<sup>-2</sup>, we observed that the source-drain current of the device increased (Fig. 1c and d), with the current of  $\sim 2.6$   $\mu$ A at  $V_{DS} = -30$  V and  $V_G = -30$  V before the proton beam irradiation increasing to  $\sim 3.1$   $\mu$ A after the proton beam irradiation at the same measurement conditions (an approximately 20% current increase). At the same time, the threshold voltage ( $V_{th}$ ) shifted to the positive gate voltage direction (see also Fig. 3a). On the other hand, when the device was irradiated with a proton beam of a higher fluence of  $10^{14}$  cm<sup>-2</sup>, the device's current decreased, with  $\sim 3.0$   $\mu$ A at  $V_{DS} = -30$  V and  $V_G = -30$  V before proton irradiation decreasing to  $\sim 2.0$   $\mu$ A after proton irradiation at the same measurement

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