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Interface engineering with double-network dielectric structure for flexible organic thin film transistors

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ABSTRACT

Taking inspiration from the characteristics and the limitations of flexible organic thin-film transistor (OTFT) devices, a novel surface modification method was proposed to modify the gate insulator to tune the mechanical compatibility and adhesion energy between gate insulator and semiconductor layer for high performance flexible OTFT devices. The surface modification method aimed to form a polymer network structure within the cross-linked polymer film by photo-polymerization of the liquid monomers which were infused into the cross-linked polymer film. The formation of double-network structure significantly improved the electrical performance of the flexible devices. The charge mobility increased from 0.17 $cm^2V^{-1}s^{-1}$ to 1.52 $cm^2V^{-1}s^{-1}$ and on/off current ratio increased by one or two orders of magnitude. During the bending tests, the devices modified with double-network dielectric structure exhibited excellent mechanical stability even after hundreds of successive bending cycles along with different bending radius. For instance, the mobility of the devices modified with methyl methacrylate only reduced by 2% after 500 successive bending cycles with a bending radius of 10 mm. It is ascribed to the improvement of mechanical compatibility and decrease of adhesion energy between gate insulator and semiconductor with the double network dielectric structure. More importantly, this work is a unique example to demonstrate the importance of mechanical compatibility and adhesion energy between gate insulator and semiconductor on the mechanical stability of flexible OTFT. Furthermore, these results clearly demonstrated that this method is a promising tool to tune the semiconductor/dielectric interface, especially the mechanical compatibility and adhesion energy between semiconductor layer and dielectric layer, which has great potential for high performance flexible OTFTs with excellent mechanical stability.

1. Introduction

Flexible electronics have attracted more and more attentions due to their promising advantages of mechanical flexibility, light-weight, low-cost and are being considered as next-generation electronics [1–3]. As one of the most promising flexible electronic devices, flexible organic thin-film transistor (OTFT) has been used in flexible displays, electronic skin, smart cards, and sensors [1,4–6]. Although flexible OTFT owns broad application prospects, the electrical performance and stability of OTFT is still needed to be improved for it commercialization, especially when the devices are under different mechanical stress [5,7,8]. Many groups have reported that mechanical bending of the devices would lead to the residual mechanical stress in the interfacial, which resulted in the formation of cracks and delamination, and affected the electrical performance and stability of flexible OTFT [5,9–13]. Therefore, eliminating the residual mechanical stress would be the key to improve the device performance and stability of flexible OTFT. Sang-Yoon Lee and coworkers demonstrated that the surface passivation of flexible devices can improve its electrical performance and stability [14]. Meanwhile, the introduction of neutral plane can effectively minimize the mechanical stress and improve the mechanical stability of flexible OTFT [15–19]. N. Yoneya and coworkers has fabricated flexible OTFT by solution process based organic semiconductor, which shown good mechanical stability after 1,000,000 times bending cycles at a radius of 5 mm [20]. Unfortunately, double-network structure has never been used as gate insulator to improve the mechanical stability of flexible OTFT.

Double-network structure has been widely used in polymer

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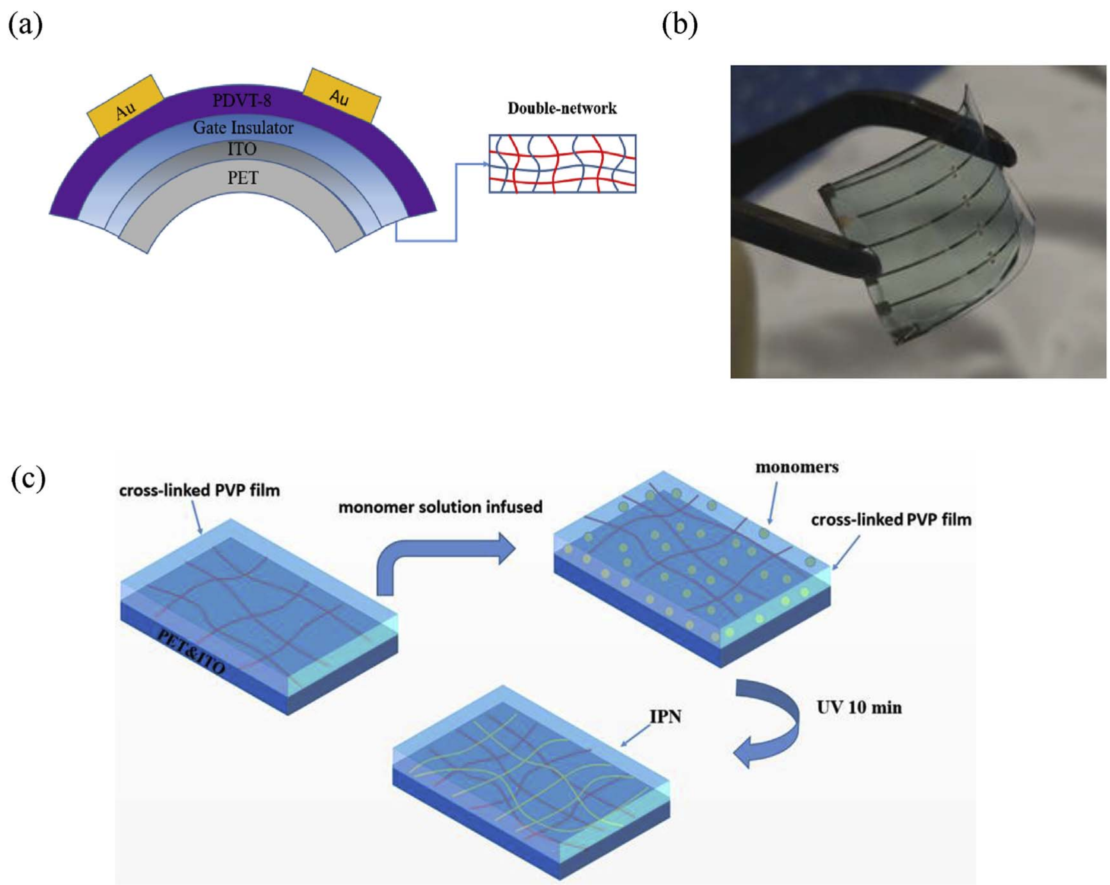

Fig. 1. (a) The schematic of flexible OTFT, (b) A photograph of flexible OTFT fabricated on the PET substrate, (c) The process of the formation of double-network structure.

elastomer and gels, which offers great freedom to tune mechanical properties, chemical resistance, and optical properties to match the intended application [21]. The definition of double-network structure is at least two kinds of intertwining polymer networks with respective peculiarities [21,22]. Each network can retain its own property without chemical bonding between two component networks, which can obvious improve mechanical strength of polymer matrix [23–25]. Besides that, the double-network structure can generate new physical and chemical surface characteristics, for example, interfacial compatibility and temperature sensitivity, and can also improve the densification of the membranes to prevent the penetration of oxygen or small organic molecule into the membranes [26–29]. As the mechanical properties of gate insulator can be modified by the double-network structure, the introduction of polymer network to the gate insulator would tune the mechanical compatibility between gate insulator and semiconducting layer, which is supposed to reduce the residual mechanical stress under bending and affect the mechanical stability of OTFT devices. Moreover, with rational selection of infused monomers, the adhesion energy between gate insulator and semiconductor layer can be optimized, which would further optimize improve the device performance.

Therefore, in this work, a novel surface modification method using double-network structure was introduced to modify the gate insulator to tune the mechanical compatibility and adhesion energy between dielectric layer and semiconducting layer of flexible OTFT. This method includes two steps: liquid monomers were infused into the cross-linked polymer films and then double-network within the surface was formed by photo-polymerization [22,30]. A key advantage of surface modification with double-network structure is that polymer network is directly infused into the gate insulator instead of blending. The surface roughness and adhesion energy of the insulator/semiconductor interface was found to be optimized with double-network structure, resulting in the improvement of carrier mobility [30–37]. More importantly, the mechanical stability of the devices was significantly improved with double-network structure as gate insulator, which is ascribed to the improved mechanical compatibility between dielectric layer and semiconductor layer. Moreover, a further improvement of device stability can be achieved with a decrease of adhesion energy between semiconductor/dielectric interface with a judicious choice of infused monomers. It showed that the charge mobility, threshold voltage and on/off current ratio of the devices modified with methyl methacrylate only slightly became worse after 500 bending cycles with a bending radius of 10 mm, while no transistor behavior was observed in the device with neat cross-linked polymer film under the same bending condition. These results clearly demonstrated that surface modification with double-network structure is a promising tool to tune the semiconductor/dielectric interface, especially the mechanical compatibility and adhesion energy between semiconductor layer and dielectric layer for high performance flexible OTFTs with excellent mechanical stability.

2. Experimental

High molecular weight π-extended Copolymer poly[2,5-bis(alkyl) pyrrolo[3,4-*c*]pyrrole-1,4(2H, 5H)-dione-*alt*-5,5′-di(thiophen-2-yl)-2,2′-(E)-2-(2-(thiophen-2-yl)vinyl)-thiophene] (PDVT-8) (Mw = 50 K, PDI = 2.4) was purchased from 1-Materials [38–40]. Poly(4-vinylphenol) (PVP) (Mw ~ 25,000), 4,40-(hexafluoroisopropylidene) diphthalic anhydride (HDA), and propylene glycol monomethyl ether acetate (PGMEA) were obtained from Sigma Aldrich. Methacrylic acid (MAA), (methyl methacrylate) (MMA) and ethylene glycol dimethacrylate (EGDMA) were received from J & K. 2, 2-dimethoxy-1, 2-diphenylethan-1-one (Irgacure 651) was obtained from BASF, and used as