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Polyurethane triblock copolymer gate dielectrics for low-voltage organic thin-film transistors

Dongkyu Kim[a], Choongik Kim[a,*], Taeshik Earmme[b,*]

[a] Department of Chemical and Biomolecular Engineering, 35 Baekbeom-ro, Mapo-gu, Sogang University, Seoul 04107, Republic of Korea
[b] Department of Chemical Engineering, Hongik University, 94 Wausan-ro, Mapo-gu, Seoul 04066, Republic of Korea

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ABSTRACT

Novel polyurethane triblock copolymers comprising polycaprolactone diol (PCL), 1,6-hexamethylene diisocyanate (HMDI), and polyethylene glycol (PEG) were synthesized for use as gate dielectric for organic thin-film transistors (OTFTs). Thin films of polyurethane gate dielectrics processed from solution exhibit excellent insulating properties (~7×10^{-7} A/cm^2 at 1 V) as well as large areal capacitance (170 nF/cm^2) with film thickness of ~50 nm. OTFTs are fabricated with representative n-channel organic semiconductor (N,N′-ditridecylperylene-3,4,9,10-tetracarboxylic diimide; PTCDI-C13) using the developed gate dielectrics, and the resulting devices show decent electrical performance with negligible hysteresis at low operating voltage of 1 V.

Introduction

In recent years, there has been increasing interest in stretchable and flexible organic electronic devices [1–16]. Among the various devices, organic thin-film transistors (OTFTs) have widely been explored as an advantageous technology to achieve stretchable transistors. To enable stretchable characteristics of OTFTs, each device component including semiconductors, conducting electrodes, and dielectric insulators should exhibit facile stretchability. Compared to prior studies of stretchable semiconductors and electrodes [17–24], research on insulating dielectric materials has relatively less explored [25–30]. High capacitance for low operating voltage, low leakage current for reliable operation, and good durability upon deformations are required for dielectric materials in stretchable OTFTs [31,32]. However, there are typically some trade-offs between the required properties. The tensile strain upon deformation can be reduced by decreasing dielectric film thickness while thin film can also result in increased leakage current [24].

Polydimethylsiloxane (PDMS), hydrogenated polystyrene-polyethylene/butylene-polystyrene (H-SEBS), or polyurethane have been reported as promising candidates for stretchable dielectrics in OTFTs with excellent insulating properties as well as robust bending durability [15,33–41]. Recently, polyurethane (PU) has attracted attention since it is easy to control the physicochemical properties of the polymer by varying co-monomer components and their ratios for better dielectric characteristics [34,37]. PU-based polymer containing a terphenyl group was reported and used as dielectrics for OTFTs [34]. The developed PU dielectric films had large film thickness (~900 nm) to reduce leakage current. Operating voltage of the device was high (~−50 V) since the dielectric constant of the developed PU-based polymer was relatively low (~4) [34]. PU and polyethylene glycol (PEG) copolymer was also reported as effective dielectric insulators in OTFTs. Although the developed copolymer exhibited high dielectric constant (~13) and decent stretchability, the film was still very thick (~1000 nm), which caused large hysteresis in the OTFT devices [37,41]. Therefore, it is desirable to develop PU-based copolymers with high dielectric constant, good insulating properties, high areal capacitance even at small film thickness for use in stretchable OTFTs. In order to improve dielectric constant of the polyurethane, free hydrogen bonded hard domains can be induced by the attached polar group [42–46].

In this paper, we demonstrate new polyurethane (PU) triblock copolymers comprising polycarprolactone diol (PCL), 1,6-hexamethylene diisocyanate (HMDI), and polyethylene glycol (PEG) as efficient dielectric insulators for stretchable OTFTs (Fig. 1). The developed triblock copolymer films with optimized monomer contents exhibited excellent insulating properties (~7×10^{-7} A/cm^2 at 1 V) as well as large areal capacitance (~170 nF/cm^2). PEG with relatively large dielectric constant (>12 at room temperature) was selected as a monomer, while PCL with high crystallinity was

* Corresponding authors.
E-mail addresses: choongik@sogang.ac.kr (C. Kim), earmme@hongik.ac.kr (T. Earmme).

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