

Structural effect on controllable resistive memory switching in donor–acceptor polymer systems

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ABSTRACT

Controllable bistable electrical conductivity switching behavior and resistive memory effects have been demonstrated in Al/polymer/indium-tin oxide (ITO) sandwich structure devices, constructed from non-conjugated vinyl copolymers of $\text{PTPA}_n\text{OXD}_m$ with pendant donor–acceptor chromophores. The observed electrical bistability can be attributed to the field-induced intra- and intermolecular charge transfer interaction between triphenylamine electron donor (D) and oxadiazole electron acceptor (A) entities, and is highly dependent on the chemical structure of the copolymers. The vinyl copolymers showed different memory behaviors, which depended on the loading of D/A ratios. The polymers containing only donor or acceptor moieties showed as insulators, the polymers containing both donor and acceptor moieties showed as WORM, flash and DRAM as D/A ratio increased. The structural effect on the physicochemical and electronic properties of the $\text{PTPA}_n\text{OXD}_m$ copolymers, viz surface morphology, thermal stability, optical absorbance and photoluminescence, and molecular orbital energy levels, were investigated systematically to study the factors that influence the memory characteristics of the devices.

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1. Introduction

With the rapid development of information technology in the 21st century, electronic products such as smart phones and tablet personal computers have become an essential part of our lives [1]. As the performance of the digital gadgets advances, the complexity of data storage device increases, with the concomitant reduction in device dimension. In order to achieve greater density of data storage and faster access to information, more components are deliberately packed onto a single chip. Existing memory technologies may survive another few generations, but are likely to approach the limit of 16 nm in 2018 [2,3], beyond which a single silicon-based memory cell becomes

less stable or reliable when storing individual bits. To maintain the Moore's Law of the cost learning curve, the potential application of molecular computation, using resistive memory switching materials made of polymers and polymer-based composites as the data storage medium, has resulted in worldwide research campaign in developing novel information storage materials and devices [4–6].

Due to the possibility of storing more data in less space and with less energy consumption, and the unique advantages of mechanical flexibility, solution processability and printability, various polymer resistive memory devices have been demonstrated recently [1,5,7–9]. In particular, donor–acceptor (D–A) polymer systems receive great attention due to their simple charge trapping/detrapping or charge transfer (CT) switching mechanism [10–14]. Moreover, the electrical function and memory characteristics of the D–A polymers can be fine-tuned over a wide

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range via versatile molecular-cum-synthesis strategy [8]. By regulating the strength of the D/A unities, the interfacial energy barrier for charge transfer and separation will be effectively adjusted, leading to electrical conductance switching phenomena in a controllable manner [13]. The loading ratio of D/A unities, which modulates the degree and stability of charge transfer interaction, defines the volatility and reversibility of the resistive memory switching in donor–acceptor polymers [12]. The packing mode of the polymer chains as well as the morphology on the interface between the D/A unities also significantly influence the charge transport behavior of the memory devices [14,15].

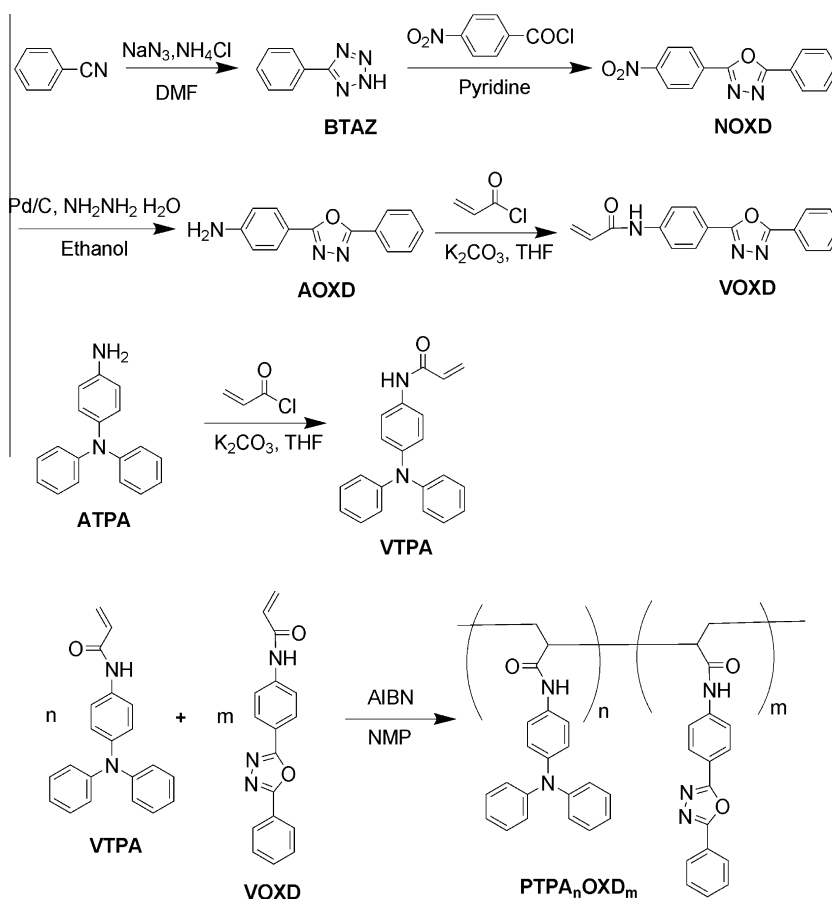
In this work, we demonstrated controllable resistive memory switching phenomena in donor–acceptor polymer systems and investigated the structure–property relationship to better understand the bistable electronic transition and memory effects. A series of non-conjugated homopolymers and random copolymers containing triphenylamine (TPA) moiety as the electron-donor and (2,5-biphenyl)-1,3,4-oxadiazole (OXD) moiety as the acceptor, which are connected to the vinyl backbone through amide linkage, were synthesized and employed as model materials to construct donor–acceptor polymer systems (Scheme 1). The morphological, thermal, optical and electrochemical

properties of the as-synthesized polymers show strong dependence on the chemical structure of the donor–acceptor systems. With continuous adjusting of the D/A loading ratios, the PTPA/POXD homopolymers and PTPA_nOXD_m random copolymers demonstrate insulating, volatile switching and non-volatile memory switching behaviors. Computation chemistry using Gaussian were performed to explore the local electronic interaction of the donor–acceptor pentamers, that are responsible for the controllable memory characteristics of the D–A polymers. The present work provides a comprehensive description on the structure-dependent electronic transition and charge transport scenario of the donor–acceptor polymer systems, and serves as guidelines for designing novel D–A polymers for high performance data storage applications and exploring the operating mechanisms of the electronic memories.

2. Experimental section

2.1. Materials

4-Nitrofluorobenzene (Acros), sodium hydride (Acros), benzonitrile (Acros), 2,2'-azobis(2-methylpropionitrile) (AIBN, Sigma–Aldrich), diphenylamine (Sigma–Aldrich), pyridine (Sigma–Aldrich), sodium carbonate (Sigma–Aldrich),



Scheme 1. Synthetic scheme of monomers and PTPA_nOXD_m copolymers.

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