

# Application of triphenylamine dendritic polymer in a complementary electrochromic device with panchromatic absorption

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## ABSTRACT

Application of poly(N, N', N''-tris[N, N-bis-(4'-diphenylamino-biphenyl-4-yl)phenyl]-1, 3, 5-benzene tricarboxamide) (PG1), a triphenylamine dendritic polymer, in an complementary electrochromic device (ECD) has been studied. Electrochromic transitions of PG1 itself give very significant transmittance change ( $\Delta T$ ) of 58.4% at 485 nm and 81.8% at 780 nm with rather good cycling stability. A complementary ECD based on the PG1 thin film and poly(3,4-ethylenedioxythiophene) (PEDOT) thin film was assembled to demonstrate panchromatic absorption characteristic through a simple fabrication process. This ECD exhibits black-to-transmissive electrochromism when switched from  $-0.4$  to  $1.5$  V. Large variation in  $L^*$  value in CIE  $L^*a^*b^*$  coordinate (45.6) and  $\Delta T$  (55.5% at 510 nm) is observed. In fact, the  $\Delta T$  value achieved in this work is comparable to the highest value (58%) among all reported triphenylamine derivative/PEDOT ECDs after an extensive literature search. Furthermore, good cycling stability is achieved in the proposed ECD, which still retained panchromatic absorption characteristic and maintained 91.7% of the initial  $\Delta T$  value at 510 nm after continuous 10,000 switchings.

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## 1. Introduction

Electrochromism is a physical phenomenon describing reversible optical absorbance change of electrochemically active materials upon an application of electrical potential [1]. In recent years, it gains a lot of attention because of its possibility in energy saving applications due to its low power consumption. Potential products such as smart windows [2–4], anti-glared rear view mirrors [5], and displays [6–9] are derived from electrochromic (EC) technologies. To make a successful product, the EC materials must have characteristics such as recognizable color changes and electrochemical reversibility [10]. Organic electrochromic polymers such as polythiophene [11,12], poly(3,4-alkylenedioxythiophene) (PXDOT) [13–20] and triphenylamine derivatives [21–23] have been studied extensively. Since the EC properties of organic polymers are tunable by tailoring their molecular structures, these materials have become popular in recent years.

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In this work, a promising anodically coloring material N, N', N''-tris [N, N-bis-(4'-diphenylamino)biphenyl-4-yl)phenyl]-1, 3, 5-benzene tricarboxamide (G1) monomer (Fig. 1) is utilized. The polymer of G1 monomer (PG1) can be effectively obtained through electro-polymerization (Scheme S1). The general aspects for electro-polymerization of triphenylamine have been extensively discussed in recent literature [24]. Moreover, the detail electrochemical reaction for polymerization of G1 has been reported in our previous work [25]. Briefly, G1 monomer would generate terminal radical cation under the applied voltage of 1.3 V (vs. Ag/Ag<sup>+</sup>). These terminal radical cations rapidly dimerize to form tetraphenylbenzidine (TPB) moiety (Scheme S1(a)) [26], which is anodically coloring and exhibits two-stage electrochromism as shown in Scheme S1(b) [27]. Therefore, the obtained PG1 film is colorless in the reduced state (bleached state). It turns red (the first colored state) when one electron per TPB group is depleted and turns deep blue (the second colored state) when two electrons per TPB group are depleted. In our previous work [28], an electrochromic device (ECD) with a large transmittance change ( $\Delta T$ ) was fabricated by utilizing PG1 film along with an ion storage layer as the counter electrode. However, the ECD does not have any absorption within the region of 600–800 nm. For practical applications, complementary ECDs, which are composed of an anodically coloring material and a cathodically coloring material [3,29–32], are desired because of their capability of harnessing two EC materials' coloring ability without increasing the consumption of electrons. And recently,

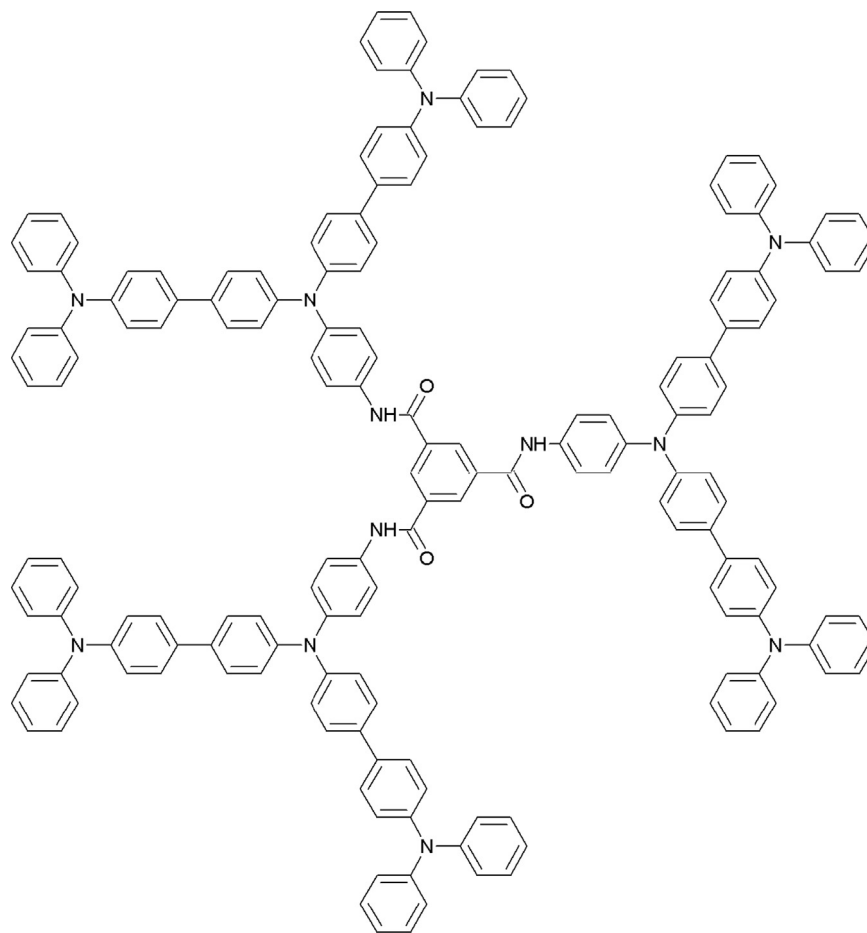


Fig. 1. Chemical structure of G1 monomer.

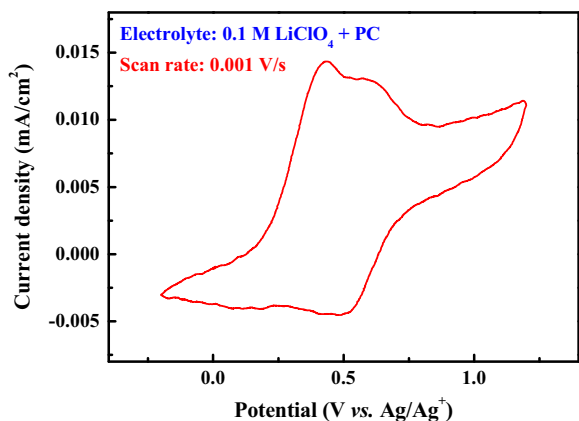


Fig. 2. Cyclic voltammetry of PG1 film scanned (scan rate: 0.002 V/s) in PC containing 0.1 M LiClO<sub>4</sub>.

black-to-transmissive ECDs become a focused issue because they are promising to be applied in emerging commercial displays like e-papers [33,34]. Reynolds et al. [35] utilized a polymer design strategy through donor–acceptor approach to successfully construct a promising panchromatic absorption electrochromic film. To eliminate the high-cost synthetic procedure proposed by the former literature [35], Kim et al. [34] fabricated a black-to-transmissive ECD by integrating four electrochromic thin films in a multi-layer configuration. Nevertheless, the visible transmittance change was still too low. Therefore, fabricating an ECD with high optical contrast and panchromatic absorption remains a challenging task [33].

Herein, a much easier approach is proposed to obtain a black-to-transmissive ECD with both bleached and colored states close to the Commission Internationale de l'Eclairage (CIE) black in  $L^*a^*b^*$  coordinate. The anodically coloring PG1 thin film is coupled with a well-known cathodically coloring poly(3,4-ethylenedioxythiophene) (PEDOT) thin film to achieve panchromatic absorption with high optical contrast. The intrinsic transparent to blue electrochromism of PEDOT can appropriately tune the color of PG1 film to render panchromatic absorption in the ECD. As the result, the proposed ECD reveals a high  $L^*$  variation ( $\Delta L^*$ ) of 45.6 in CIE  $L^*a^*b^*$  coordinate and a high  $\Delta T$  of 55.5% at 510 nm. Moreover, 91.7% of the initial  $\Delta T$  is maintained after 10,000 continuous switching cycles.

## 2. Experimental

### 2.1. Materials

*ortho*-Dichlorobenzene (ODCB, 99.7%) was purchased from TEDIA. LiClO<sub>4</sub> (95%), tetrabutylammonium perchlorate (TBAP, > 99%) and propylene carbonate (PC, 99%) were all purchased from Alfa Aesar. The G1 monomer (> 99%) was synthesized according to the previous work [25]. The PC solution was dehydrated at least overnight by using 4-Å molecular sieves from Acros. All the other reagents were used as received without further purification.

In this study, both PG1 and PEDOT electrochromic films were electrodeposited on the substrate of indium-tin oxide (ITO) conducting glass (Solaronix SA,  $R_{sh}=7\ \Omega/\square$ ) using a three-electrode system, where ITO electrode was used as the working electrode,

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