



# Effect of imidazole derivatives in triphenylamine-based organic dyes for dye-sensitized solar cells



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## ABSTRACT

In order to study the influence of imidazole derivatives in triphenylamine-based organic dyes, two different imidazole derivatives are introduced into the phenyl ring of the triphenylamine core, coded as **TPA-B5** and **TPA-B6**, respectively. The photophysical and electrochemical properties of the dyes are investigated by UV–vis spectroscopy and cyclic voltammetry. **TPA-B5** increases the molar extinction coefficients and  $\lambda_{\max}$  because of the extension of the  $\pi$ -conjugation structure of the dye and non-planar structure of imidazole derivative. However, **TPA-B6** does not increase the molar extinction coefficients and  $\lambda_{\max}$  compared with a simple triphenylamine derived dye (**TPA-1**), which may be due to the planar structure of imidazole derivative and benzene ring. The structure of **TPA-B6** is in favor of the formation of dye aggregates on the semiconductor surface and the recombination of conduction band electrons with triiodide in the electrolyte. Overall conversion efficiencies of 3.13% and 1.21% under full sunlight (AM 1.5G, 100 mW cm<sup>-2</sup>) irradiation are obtained for DSSCs based on the two new dyes, under the same conditions, the dye **TPA-1** and ditetrabutylammonium cis-bis(isothiocyanato) bis(2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium(II) (**N719**) give overall conversion efficiencies of 2.23% and 5.38%, respectively. Although the overall conversion efficiencies of these dyes are not very high, the results will still afford significant value for future development of efficient D– $\pi$ –A sensitizers.

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## 1. Introduction

Along with increasing energy demands and global warming problems, dye-sensitized solar cells (DSSCs) have received more and more attention due to their relatively high power conversion efficiency and low cost alternative to the conventional photovoltaic devices [1,2]. As a key part of DSSCs, the dyes play a significant role for the DSSCs. The ruthenium dyes produce overall photo-electrical conversion efficiency of higher than 10% due to their broad photoresponse area [3–5]. However, the manufacturing costs and environmental issues of the ruthenium sensitizers limit their large-scale application. Organic dyes have

advantages as photosensitizers due to their high molar extinction coefficients, tuning of absorption wavelength, and relatively facile design and synthesis. To find new organic dyes with cheaper cost and higher efficiency, lots of efforts have been devoted. Zinc porphyrin dye has been reported with a high photo-electrical conversion efficiency ( $\eta$ ) exceeding 12.0% under standard AM 1.5 sunlight irradiation [6]. On the other hand, the performance of DSSCs based on metal-free organic sensitizers has recently been improved, the photo-electrical conversion efficiency up to 10% values has been reported [7–10].

In metal-free organic sensitizers, the traditional linear donor– $\pi$ –acceptor (D– $\pi$ –A) system has the most common character, which is consisted of donor,  $\pi$ -conjugation linker, and acceptor part. But the D– $\pi$ –A structure usually has a linear construction, and the major factors for low

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conversion efficiency of DSSCs based on organic dyes are the formation of dye aggregates on the semiconductor surface and the recombination of conduction band electrons with triiodide in the electrolyte [11]. In order to overcome the disadvantages and extend the  $\pi$ -system of the dye, introduce an imidazole derivative to the D- $\pi$ -A structure, which is possible alternative to retard the interfacial charge-recombination dynamics and to retain efficient light-induced charge separation.

Among the organic dyes, triphenylamine (TPA) and its derivatives as donor units have displayed promising properties in the development of photovoltaic devices, because the TPA molecule is non-coplanar, chemical stability and can act as rich electron donor [12–14]. A series of organic dyes showed that extending the  $\pi$ -conjugation could improve the photovoltaic performance of the DSSCs [15–18]. So based on those studies, we introduce the imidazole derivatives to the structure of TPA, and the cyanoacetic acid act as acceptor group to constitute the D- $\pi$ -A structure. We therefore designed and synthesized two new dyes (TPA-B5 and TPA-B6) with different imidazole derivatives. At the same time, the corresponding dye (TPA-1) and N719 act as the purpose of comparison dyes, which were successfully applied in DSSCs. The effects of the different imidazole derivatives in TPA-B5 and TPA-B6 on the optical, electrochemical properties and photovoltaic performances were studied. The corresponding molecular structures of the three dyes and N719 are shown in Scheme 1.

## 2. Experimental section

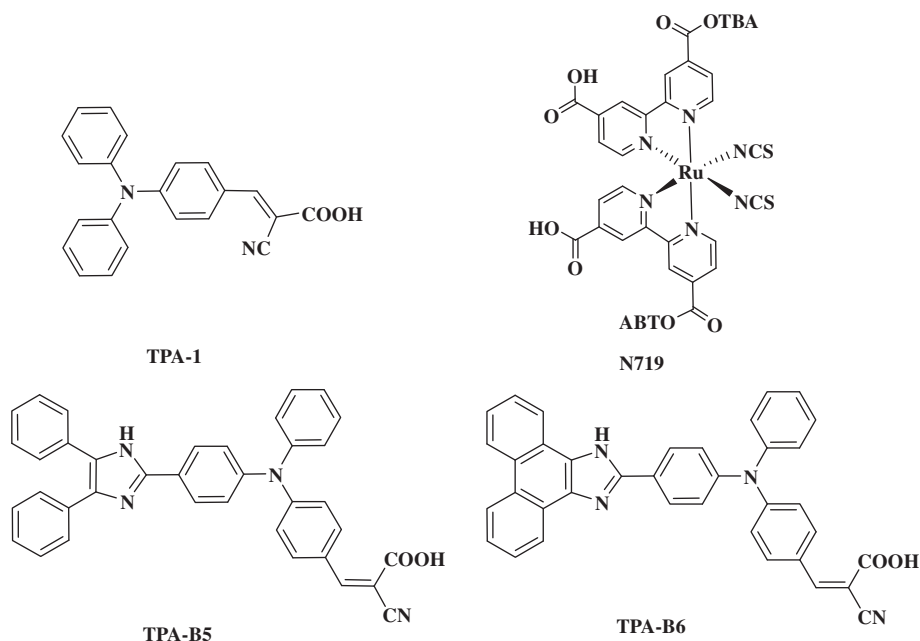
### 2.1. Materials and characterization

All solvents and other chemicals were reagent grade and used without further purification. 1,2-diphenyleth-

ane-1,2-dione, phenanthrene-9, 10-dione, cyanoacetic acid and triphenylamine were purchased from Astatech. HRMS data were obtained with a HITACHI-80 instrument.  $^1\text{H}$  NMR spectra were measured on Varian Mercury Plus 400 MHz NMR instrument. Mp data were obtained on X4 melting point detector (FUKA, Beijing, China). INSPECT F Scanning electron microscopy (SEM) (FEI, Netherlands) was used to measure the thickness of the  $\text{TiO}_2$  film. Absorption spectra were measured with SHIMADZU (model UV2550) UV-vis spectrophotometer. Cyclic voltammetry experiments were performed on a CH Instruments 660C electrochemical workstation with a scanning rate of 50 mV/s in dimethylformamide (DMF) ( $5.0 \times 10^{-4}$  M) containing 0.1 M  $n\text{-Bu}_4\text{NPF}_6$  as the supporting electrolyte, platinum as counter and work electrodes and Ag/AgCl as reference electrode.

### 2.2. Fabrication of DSSCs

$\text{TiO}_2$  colloid was prepared according to the literature [19]. The washed FTO glass substrates were immersed in 40 mM  $\text{TiCl}_4$  aq. at 70 °C for 30 min to form a compact layer of  $\text{TiO}_2$ , which plays an important role in suppressing the charge recombination of DSSCs at the interface between FTO and electrolyte, then washed with water and ethanol. A thin film of  $\text{TiO}_2$  was prepared on the FTO substrate with the compact  $\text{TiO}_2$  layer through blade coating with glass rod. After drying the nanocrystalline  $\text{TiO}_2$  layer at 80 °C, the  $\text{TiO}_2$  thin film with more layers was achieved by repeating the blade coating above process two times. The resulting working electrode was composed of a 15  $\mu\text{m}$  thick transparent  $\text{TiO}_2$  nanoparticle layer. Finally  $\text{TiO}_2$  electrodes were treated at 450 °C for 30 min. After cooling to room temperature, the electrodes were immersed in 40 mM  $\text{TiCl}_4$  aq. at 70 °C for 30 min, and washed with



Scheme 1. Molecular structures of TPA-1, N719, TPA-B5 and TPA-B6.

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