



# Novel planar binuclear zinc phthalocyanine sensitizer for dye-sensitized solar cells: Synthesis and spectral, electrochemical, and photovoltaic properties



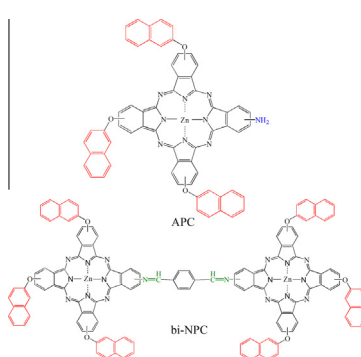
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## HIGHLIGHTS

- APC and bi-NPC were designed for use in dyes-sensitized solar cells.
- Bi-NPC was synthesized based on Schiff base.
- Schiff base could form coplanar condition between mononuclear phthalocyanines.
- Bi-NPC shows the higher absorbance, the better red shift and performance.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 25 June 2014

Received in revised form 1 September 2014

Accepted 11 September 2014

Available online 19 September 2014

### Keywords:

Planar binuclear

Dye-sensitized solar cells

Schiff base

Electrochemistry

## ABSTRACT

A planar binuclear zinc phthalocyanine was newly synthesized for use in dye-sensitized solar cells, based on Schiff base and asymmetric amino zinc phthalocyanine. The novel compounds were characterized using FTIR, UV–Vis,  $^1\text{H}$  NMR, cyclic voltammetry and elemental analysis. From the reduction and oxidation behavior, it is proved that APC and bi-NPC have negative LUMO levels and positive HOMO levels, satisfying the energy gap rule, and can be employed as sensitizers for dye-sensitized solar cells (DSSCs) applications.

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## Introduction

As one of the most promising methods for future low cost power production from renewable energy sources, DSSCs have the outstanding advantage of high light-to-electricity conversion efficiencies and easy fabrication [1]. The absorption properties of dyes dictate the light-harvesting capacity of DSSCs, since the light absorption function is fulfilled by the dye and the electron and hole

transportation are fulfilled by the nanocrystalline metal oxide and electrolyte. As a kind of functional material, the dye should be soluble in solvents that are compatible with the  $\text{TiO}_2$  and favorable for absorption of a nonaggregated monolayer on the surface [2]. Phthalocyanine is a special dye pigment consisting of 18- $\pi$  electronics and has a similarity with biological molecules chlorophyll and hemoglobin [3,4]. Strong Q band light absorption properties at around 700 nm and excellent chemical stability make phthalocyanine become a better choice as sensitizer using in DSSCs. In addition, as the characteristic of color is bright and structure could be modified, phthalocyanine is widely used in different areas, such

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as photographic printing [5], photovoltaic cells [6–8], liquid crystals [9,10], photodynamic therapy [11–14], light-emitting diodes [15,16] and chemical sensors [17,18]. Phthalocyanines have various central metals and extended  $\pi$ -conjugation system and have been tested several decades as sensitizers of wide-band gap oxide semiconductors, as increasingly high incident photon-to-electric current conversion yields are obtained [19–23]. However, poor solubility of macrocycle in organic solvent, strong tendency to aggregation on the film surface, lack directionality in the excited state and easy electron recombination between injected electron in  $\text{TiO}_2$  conduction band and oxidized dye hinder the incident photon-to-electric current conversion yields [2].

This paper concerns phthalocyanines in which polar were attached to the periphery together with an additional group to enhance solubility in organic solvents and Schiff base connect two asymmetric mononuclear phthalocyanines. A new planar binuclear zinc phthalocyanine (9(10), 9'(10'), 16(17), 16'(17'), 23(24), 23'(24')-( $\beta$ -naphthoxy)-2(3), 2'(3')-(1,4-methanamide phenyl) zinc phthalocyanine (bi-NPC)), based on Schiff base and extended  $\pi$ -conjugation concept connecting two mononuclear unsymmetrical phthalocyanines, and asymmetric amino zinc phthalocyanine (9(10), 16(17), 23(24)-( $\beta$ -naphthoxy)-2(3)-(amido) Zinc Phthalocyanine (APC)) have been designed and synthesized for the dye-sensitized solar cells (Fig. 1). APC has three  $\beta$ -naphthoxy groups to enhance the solubility of phthalocyanine in common organic solvent, reduced the aggregation and turned the LUMO level of phthalocyanine. Bi-NPC is made of two unsymmetrical amino zinc phthalocyanines connected on Schiff base reaction. Formyl groups of 1, 4-phthalaldehyde and the amino of asymmetric mononuclear phthalocyanines form the Schiff base. Schiff base could provide a coplanar condition between two mononuclear phthalocyanines [24]. As reagents in organic synthesis and liquid crystal materials, Schiff base compounds and their metal complexes containing imine or methylene amine groups ( $-\text{RC}=\text{N}-$ ) have important application in medicine, catalysis, analytical chemistry, corrosion and photo

chromic. Compared with general method of ordinary mononuclear phthalocyanine by changing substituent to enhance conjugate system, C=N bond of Schiff base connecting two mononuclear phthalocyanines could extend conjugation between the two mononuclear unsymmetrical phthalocyanines. To a certain extent, it integrates and arranges the phthalocyanine molecules.  $\beta$ -naphthoxy could increase the bi-NPC solubility in DMF, on the other hand reduce accumulation between the phthalocyanine molecules based on the space steric hindrance. The synthesis and spectral, electrochemical and photovoltaic properties of these phthalocyanines based sensitizers are investigated.

## Experimental

4-Nitro-phthalonitrile, 1,4-phthalaldehyde,  $\beta$ -naphthol, 1,8-diazabicyclo[5,4,0]-undec-7-ene (DBU), 1-pentanol, N,N-Dimethylformamide (DMF), tetrahydrofuran (THF),  $\text{Na}_2\text{S}\cdot 9\text{H}_2\text{O}$ ,  $\text{CHCl}_3$ , HAC, anhydrous  $\text{K}_2\text{CO}_3$ , anhydrous  $\text{MgSO}_4$ , anhydrous  $\text{SiO}_2$  were purchased commercially. DMF, THF and  $\text{CHCl}_3$  were dried and distilled by accustomed methods before use. All other solvents and chemicals used in this work were analytical grade and used without further purification. Column chromatography was performed on silica gel (200–300).

## Synthesis

### 4-( $\beta$ -Naphthoxy) phthalonitrile (1)

Phthalonitrile derivative was prepared by similar method reported in the literature [25,26]. 4-Nitro-phthalonitrile (1.732 g, 0.01 mol) and  $\beta$ -naphthol (1.442 g, 0.01 mol) were added successively with stirring to dry DMF (30 ml). After dissolution, anhydrous  $\text{K}_2\text{CO}_3$  (2 g, 0.014 mol) was added and the reaction mixture was stirred at 60 °C. Further anhydrous  $\text{K}_2\text{CO}_3$  (1 g, 7 mmol) was added portion-wise after 2 h. Stirring vigorously for 28 h under nitrogen. Then the reaction mass was poured into 200 ml of cold

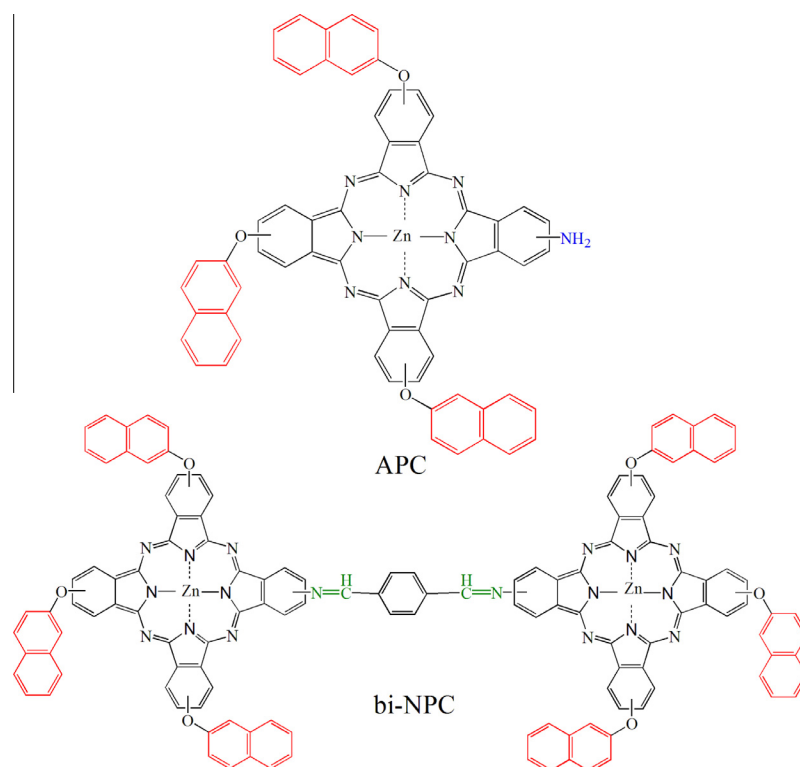


Fig. 1. The molecular formula of APC and bi-NPC.

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