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Effect of donor-donor- π -acceptor architecture of triphenylamine-based organic sensitizers over TiO_2 photocatalysts for visible-light-driven hydrogen production

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ABSTRACT

In this paper, we report a series of triphenylamine (TPA) donor based dyes in sensitized hydrogen production over TiO_2 –Pt catalysts under visible light. Among the selected dyes, 4-(diphenylamino) phenylcyanoacrylic acid (DN-F01), 5-[4-(diphenylamino)phenyl]thiophene-2-cyanoacrylic acid (DN-F02), 3-(5-(4-(diphenylamino)styryl) thiophen-2-yl)-2-cyanoacrylic acid (DN-F03), (E)-3-(5-(4-(bis(2',4'-dibutoxy-[1,1'-biphenyl]-4-yl)amino)phenyl) thiophen-2-yl)-2-cyanoacrylic acid (DN-F04) and 3-(6-(4-(bis(2',4'-dibutoxy-[1,1'-biphenyl]-4-yl)amino)phenyl)-4,4-dihexyl-4H-cyclopenta[1,2-b:5,4-b'] dithiophen-2-yl)-2-cyanoacrylic acid (DN-F05) were adsorbed on Pt– TiO_2 and tested for photocatalytic hydrogen production under visible-light in presence of triethanolamine (TEOA) as sacrificial electron donor (SED). The enhanced light absorption and effective interfacial charge transfer from excited dye to TiO_2 is significant in DN-F05– TiO_2 . The DN-F05 sensitized Pt– TiO_2 (DN5@T) photocatalyst exhibited higher turnover number (TON 1864) and apparent quantum efficiency (AQE ~44%) when compared to their corresponding simple architecture dye molecules. Moreover, the additional donating group di-butoxyphenyl and dihexyl-cyclopentyl π -conjugated bridge units provide an excellent surface protection through steric hindrance leading to a good photocatalytic performance. Electrochemical and computational studies suggest the better photocatalytic activity of DN5@T. The photocatalyst shows long term stability and reproducibility at neutral pH.

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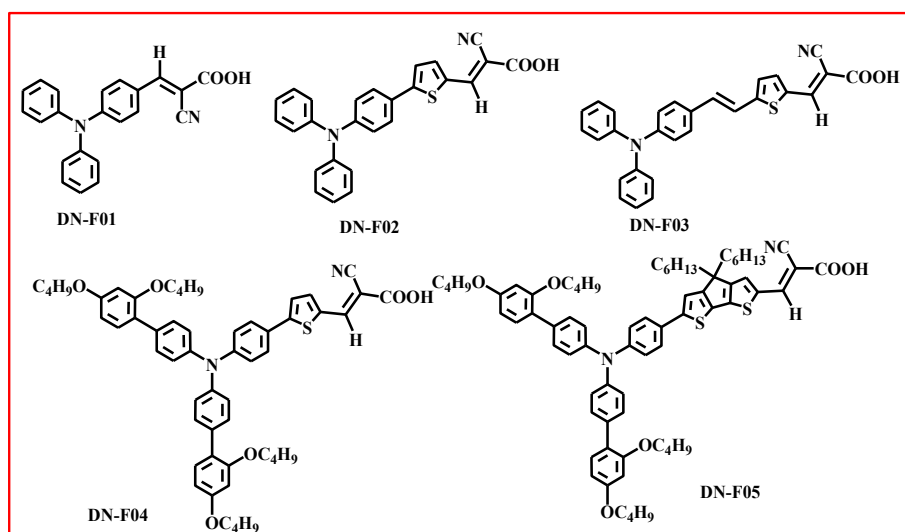
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Introduction

In the past three decades, enormous attention has been focused on the development of active photocatalysts in the production of hydrogen from water and renewable energy sources which could lead to the generation next clear and green fuel [1,2]. Taking into account a large number of patents and scientific literature published on hydrogen generation using semiconductors as the photoactive components in the systems. The majority of photocatalysts are, however wide band-gap semiconductors which are active only under UV irradiation. TiO_2 is most widely used semiconductor despite its limited light harvesting ability. Dye sensitization proved to be one of the powerful strategies for dealing with this issue [3–7]. After the pioneering success achieved by O'Regan and Gratzel in 1991 [8], dye-sensitized solar cell and dye-sensitized photocatalytic reactions have attracted ever increasing attention [9,10]. Up to now, metal complexes such as Ru or Pt [11–13], metal complex porphyrins [14,15], phthalocyanines [16]. However, in metal complex sensitizers, they have encountered problems, such as limited resources, high synthetic cost and difficult purification and environmental toxicity. On the other hand, organic dyes as photosensitizer are inexpensive and diverse in forms which show much stronger light-harvesting ability than metal complexes. In particular, eosin Y [17,18] and Rhodamine B [19] have been widely used as sensitizers for hydrogen production under visible-light. While concerning the dye sensitized photocatalytic hydrogen production, several reports can be found to enrich the state-of-art. Abe et al. have described an overall water splitting system with two-step photoexcitation using commercial coumarin and carbazole based sensitizer [20,21]. T. Ishihara et al. have properly explained the spacer effects in metal-free organic sensitizer which achieved high TON 4460 for hydrogen production after 27 h [22]. Phenothiazine-based organic dyes with two anchoring groups were also implemented for photocatalytic hydrogen generation on TiO_2 semiconducting system [23], Xanthene dyes have also been

displayed high efficiencies in photocatalytic system [3,24–26]. Furthermore, in recent past, different dye sensitized systems were developed where TiO_2 has not been used as the semiconductor [27,28]. However, their AQE are not high enough in the visible region. There are still some challenging issues such as dye stability, dye desorption and recombination during the photoreaction. Therefore, more exploration for durable and efficient dyes is required.

Organic dyes with triphenylamine (TPA) groups as a donor and cyanoacrylic group as acceptor or anchoring group exhibit promising properties and have attracted much attention in dye sensitized solar cell [29–32]. The organic dyes with triphenyl moieties possess unique characteristics like (i) non planarity, (ii) high molar absorption coefficients, (iii) fine tunable energy levels (HOMO and LUMO); (iv) low dye aggregation. However, these properties led to low recombination and back-reaction in the solar energy conversion process. Moreover, the π -conjugated system of the sensitizing molecule could be modified to obtain absorption spectra which are determined by the band gap of molecules. Owing to these desirable molecular properties, TPA based sensitizers are highly acceptable in the application of DSSCs, A. Hagfeldt et al. have described the bulky di-butoxyphenyl substituted electron donating groups on the TPA provide an excellent surface protection through steric hindrance, leading to a good photovoltaic performance independent of the presence of coadsorbent during dye adsorption [33]. X. Yang et al. developed the intramolecular energy transfer and intramolecular charge transfer properties of TPA dyes [34]. In this study, among the selected TPA sensitizers, their molecular structures are displayed in Scheme 1, in which namely, DN-F01 is smaller in size and spacer free. Apart from this, all the other dyes have three main components; a donor part (D), a linker (L) and an acceptor group (A). An extended π -conjugation with a methine chain in DN-F03 would probably give a red shift of the absorption peak with respect to DN-F02 and DN-F01. DN-F04 and DN-F05 sensitizers possess D- π -A groups along with additional o- and p-substituted four butoxy groups. In the DSSCs report, the



Scheme 1 – Molecular structure of dyes.

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