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Shining light on panchromatic ruthenium sensitizers towards dye-sensitized photocatalytic hydrogen evolution

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ABSTRACT

Five new photocatalysts have been synthesized in order to extend the photo response upto visible range, by adsorbing MC113-MC117 ruthenium complexes on TiO₂-Pt composites. Highlight harvesting properties of these ruthenium complexes instigated us to evaluate for photocatalytic activity. The absorption curves of the synthesized photocatalysts extended up to 750 nm. Morphological studies of photocatalysts have been carried out using SEM and powder X-ray crystallography. Among all photocatalysts, MC113PC showed high photocatalytic activity i.e. 9474 TONs. IPCE and fluorescence quenching studies of the catalysts revealed the light harvesting nature and electron injection efficiency. The photocatalytic activity of MC photocatalysts were systematically screened at different pH and employing different sacrificial electron donors (SED) in order to obtain optimal photocatalytic performance.

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Introduction

Photocatalytic hydrogen generation by water splitting has attracted extensive research interest towards achieving green globalisation and to develop a society that uses a cleaner fuel sources. Honda–Fujishima were the first to introduce the water splitting effect using a rutile TiO₂ and a Pt black system in 1972 [1]. Thereafter a large number of research groups have

shed focus on the synthesis of photocatalysts for water splitting process under UV irradiation using wide-band gap semiconductors such as TiO₂ [2–4]. Unfortunately, the visible light active materials are not thoroughly investigated for this purpose and still have large space for development [5–9]. One of the most widely used and popular photocatalyst TiO₂ is inactive in the visible range because of its large band gap (ca.3.2 eV) thereby limiting its practical applications. Among the various routes for visible light exploitation, dye

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sensitization of TiO_2 [10–15] is a viable and widely used strategy that has been extensively studied and found to be successful in dye sensitized solar cells (DSSC) [16–20]. According to this strategy dye molecule absorbed on the TiO_2 surface undergo photo excitation and injects the electrons into the conduction band of TiO_2 , thereby generating photocurrent through external circuit. This strategy has been successfully employed for the hydrogen production too from sensitized TiO_2 . Although developments are blooming in this field, extensive studies for more efficient systems are warranted. Among several transition metals, metalloporphyrins [21], phthalocyanines [22] and Ruthenium dyes [23] have displayed high efficiency for visible-light-induced hydrogen production from water. This is presumed to be because of their long excited state lifetime. In contrast, $\text{Ru}_2(\text{bpy})_4(\text{BL})(\text{ClO}_4)_2$ (bpy = bipyridyl, BL = bridging ligand) complex showed unique “antenna effect” to capture more visible light for remarkable hydrogen production over dye-Pt/ TiO_2 system [24,25]. TiO_2 and ZnO sensitized with $[\text{Ru}(\text{dcbpy})_2(\text{dppq})]^{2+}$ proved to be an extremely stable and efficient photocatalytic system for hydrogen evolution from water even in the absence of an electron donor under visible light [26]. Carboxylic acid anchoring group is the most widely used functional group for grafting on the TiO_2 metal surface, owing to ready accessibility from commercially available precursors. However, carboxylic acid anchoring groups are prone to dissociate from the TiO_2 surface under alkaline electrolyte conditions. In order to improve the hydrolytic stability of dye on TiO_2 , Reisner and coworkers [27,28] tested photocatalytic activity with phosphonic acid anchoring groups and demonstrated their better hydrolytic stability over carboxylic acid. However, phosphonic acid anchoring groups suffer from low electron injection efficiency onto the TiO_2 conduction band compared to the carboxylic acids, due to the sp^3 hybridisation

of phosphonic acid, which hindered the delocalization over the anchoring [29,30]. However investigation of robust anchoring group for dye-sensitized photocatalyst is necessary, further major challenge is to tune the spectral properties of these sensitizers to improve their light harvesting capability.

Panchromatic sensitization by heteroleptic Ru complexes attracted intensive interest in the past few years for different photocatalytic applications [31]. Properties inherited by these complexes like strong absorption in the visible region, long excited state lifetime, favourable energy levels for reduction of catalysts and oxidation of sacrificial agent adds to their versatile applications [32,33]. Especially, thiocyanate terpyridine based ruthenium complexes are well-known for panchromatic light harvesting nature. In this regard, our group has been actively involved in the synthesis of sensitizers for DSSC application and achieved record efficiency for a device employing co-adsorbent technology with panchromatic terpyridine ruthenium complex as a base dye [34]. Subsequently, we have synthesized five novel 4'-functionalised terpyridine based ruthenium dyes that revealed good panchromatic absorption for DSSCs [35]. However, among several ruthenium complexes employed for the photocatalytic water splitting, very few were reported with suitable optical properties λ_{max} 580 nm and tailing up to near IR region (750 nm) on TiO_2 [32,33,47]. Inspired by the role of sensitizers in visible light driven dye-sensitized photocatalysis for efficient hydrogen production, we herein employed five novel 4'-functionalised terpyridine based ruthenium dyes (Fig. 1) as efficient photocatalysts to produce hydrogen from water using triethanolamine (TEOA) as sacrificial electron donor (SED). As per our knowledge, this is by far the first report in the study of thiocyanate terpyridine based ruthenium complexes as photocatalysts.

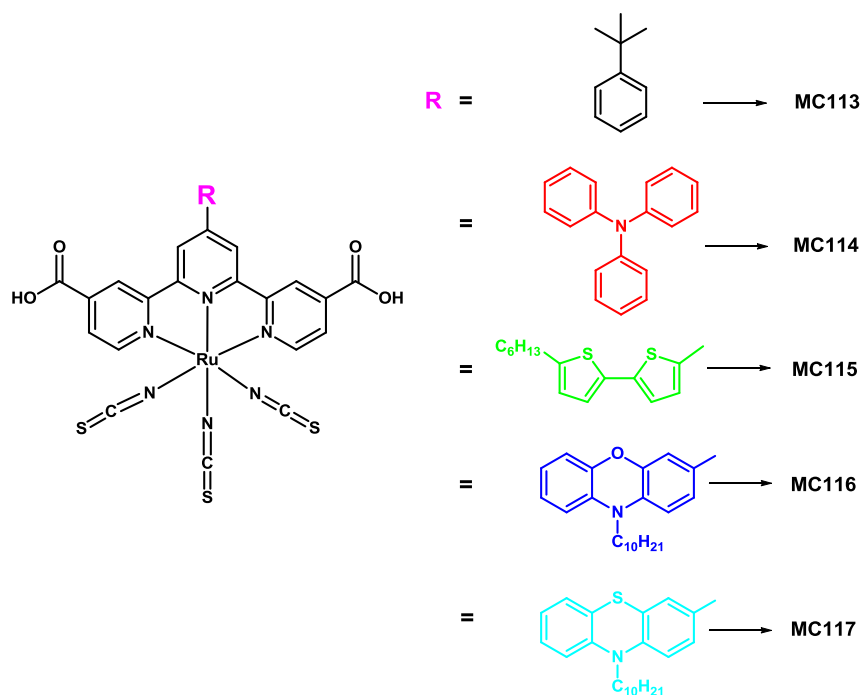


Fig. 1 – Structures of the MC dyes.

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