



Novel ruthenium sensitizer with multiple butadiene equivalent thienyls as conjugation on ancillary ligand for dye-sensitized solar cells



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ABSTRACT

A heteroleptic polypyridyl ruthenium complex 'cis-Ru(4,4'-bis(3,5-bis(5-hexylthiophen-2-yl)phenyl)-2,2'-bipyridine)(4,4'-dicarboxyl-2,2'-bipyridine) (NCS)₂, **MC102'**, with a high molar extinction coefficient was synthesized and characterized with IR, ¹H NMR, Mass, UV–Vis spectroscopy. The test cell DSSC devices constructed with 0.23 cm² active area photo-electrode in combination with an electrolyte composed of 0.6 M dimethylpropyl-imidazolium iodide (DMPII), 0.05 M I₂, and 0.1 M LiI in acetonitrile yielded solar to electric energy conversion efficiency (η) of 4.42% under Air Mass (AM) 1.5 sunlight, while the reference **N719** sensitized solar cell fabricated and evaluated under similar conditions exhibited η -value of 5.84%.

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1. Introduction

Dye sensitized solar cells (DSSCs) have attracted special attention due to their efficient performance, ease of fabrication and economy of production [1]. Recently much effort has been devoted to the synthesis and characterization of various sensitizers, such as ruthenium complexes, zinc porphyrins and metal free organic dyes for DSSCs [2,3]. The sensitizer on absorption of photon from sunlight injects an electron onto the TiO₂ conduction band from its excited state and the charge separation takes place at the interface of sensitizer/TiO₂. In order to obtain maximum efficiency, the electron and hole should be transported towards opposite electrodes by TiO₂ particulate network and the electrolyte respectively as fast as possible before the

recombination occurs. To control the recombination and hence to improve the efficiency of the device, several new sensitizers, TiO₂ architectures and efficient electrolytes have been developed. However the devices constructed from ruthenium polypyridyl complexes such as **N3** and **N719** dyes, nano particle TiO₂ and liquid electrolytes (Iodine/triiodide) attained power conversion efficiency of >10% under one-sun illumination [2d].

Cobalt (II/III) based redox electrolytes are found to be most efficient in porphyrin-sensitized devices which exhibited efficiency that exceeds 12%, highest to date [4]. Recently Brian O'Regan achieved 4% efficiency in a DSSC using water/iodide electrolyte system [5]. To explore new DSSC architectures, energy relay dyes (ERDs) attached with the sensitizing dyes (SDs) or coadsorbed on TiO₂ surface have been experimented to improve overall efficiency of the device via efficient Forster resonant energy transfer from ERD to SD [6]. A light absorbing conjugated polymer, poly(3-hexylthiophene) (P3HT) has been effectively used

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as both ERD and hole transporter in a novel solid-state DSSC device structure [6f]. Improving the performance of the device without addition of new element that would otherwise demand further optimization is an important issue and hence N719 modification is still an attractive alternative. While the anchoring dicarboxy bipyridine grafts the sensitizer over the semiconducting titania surface for efficient electronic communication, the other ancillary bipyridine can be structurally modified to tune the spectral and electronic properties of the sensitizer molecule to obtain optimum light to electricity conversion efficiency.

Since the publication of reports on HRS1 [7a] and C101 [7b] as efficient sensitizers for DSSC applications, thiophene bridges played major role in ruthenium as well as in metal free organic sensitizers to enhance the light harvesting ability [8]. Pendent alkyl thiophenes attached directly or conjugated with double/triple bonds on ancillary bipyridine ligand of ruthenium complexes have been reported [7a,b,8f].

Ruthenium complexes synthesized in our laboratory having *meta* di *tert*-butyl phenyl moieties substituted directly (H101) [9a] and with conjugation (HRD1) [9b] on ancillary bipyridyl proved to be excellent sensitizers for DSSC applications. As a consequence we constructed a hybrid of K19 [2i] and HRD1 sensitizers to integrate the properties of both the molecules in one sensitizer, MC119 [9c] and obtained better light harvesting properties with conversion efficiency of 8.36%. Thiophene is a stable 5-membered heterocyclic ring system where two double bonds are in conjugation (butadiene) and are locked with an electron rich sulfur atom providing aromatic stability and facilitates functionalization at other positions. Taking advantage of these features we designed a new sensitizer 'cis-Ru(4,4'-bis(3,5-bis(5-hexylthiophen-2-yl)phenyl)-2,2'-bipyridine)(4,4'-dicarboxyl-2,2'-bipyridine) (NCS)₂ which has four alkyl thiophenes on the ancillary ligand of the ruthenium complex coded as MC102 (Fig. 1). The advantages of the sensitizer include: (1) the four alkyl chains increase the solubility while preventing direct contact of the electrolyte with the TiO₂/conducting glass [10] (2)

thiophenes with high conjugation increase the molar extinction coefficient and red shift the metal to ligand charge transfer absorption band while providing thermal and photo stability and (3) phenyl rings act as spacers which support the two alkyl thiophenes at *meta* positions. Thus overall features of this sensitizer are promising to enhance the light harvesting power of the sensitizer. In this paper, we synthesized and characterized the new ruthenium sensitizer MC102 and fully characterized with IR, ¹H NMR, Mass, UV–Vis spectroscopic measurements apart from evaluating the electrochemical properties and the photovoltaic properties and compared with that of standard N719 sensitizer.

2. Materials and methods

2.1. General

Cyclic voltammetric measurements were performed on a PC-controlled CH instruments model CHI 620C electrochemical analyzer and the experiments were performed on 1 mM dye solution in acetonitrile at scan rate of 100 mV s⁻¹ using 0.1 M tetrabutyl ammoniumperchlorate (TBAP) as the supporting electrolyte. The working electrode was glassy carbon, the standard calomel electrode (SCE) was used as the reference electrode and platinum wire was an auxiliary electrode.

2.2. Fabrication of test cells

The test cells were fabricated using a sintered TiO₂ plate (at 500 °C for 30 min followed by cooling to 80 °C) which was immersed into the dye solution at room temperature for 24 h. The dye solution contained 0.2 mM MC102 in acetonitrile and *tert*-butyl alcohol (1:1). The counter electrode was prepared by coating an FTO plate with a drop of H₂-PtCl₆ solution (2 mg of Pt in 1 mL of ethanol) and heating it at 430 °C for 15 min. The dye-coated TiO₂ electrode and Pt counter electrode were assembled into a sealed

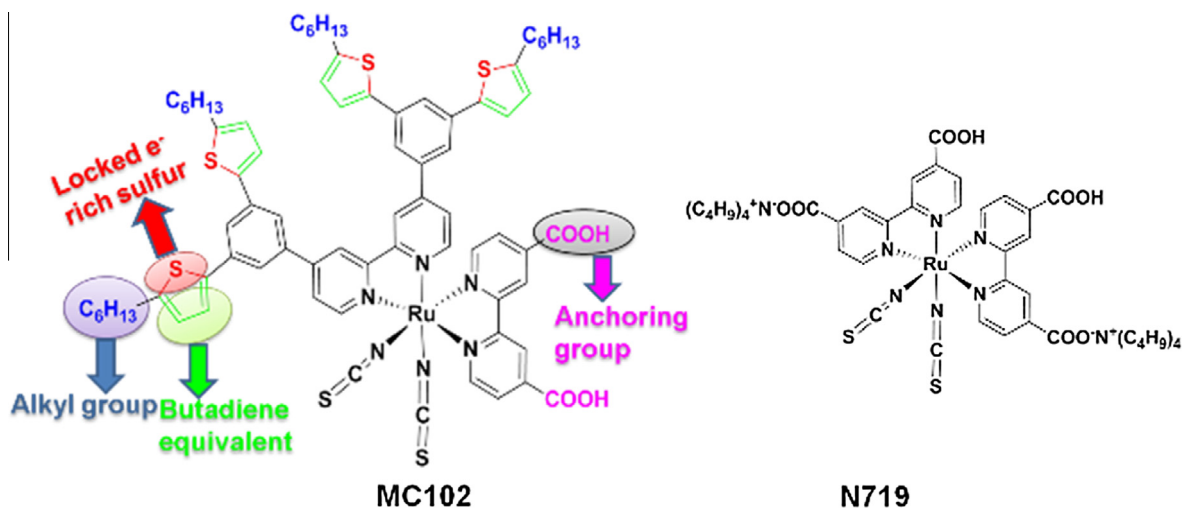


Fig. 1. Molecular structures of the MC102 and N719 sensitizers.

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