

## Efficient quasi-solid-state dye sensitized solar cells based on a novel ruthenium sensitizer

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### ABSTRACT

A new sensitizer, Ru(2,2'-bipyridine-4,4'-dicarboxylic acid)-4,4'-bis(2-(2,4,6-tri-methoxyphenyl)ethenyl)-2,2'-bipyridine) (NCS)<sub>2</sub>, denoted K77-3, was synthesized. The UV–vis spectrum of K77-3 was characterized. Dye sensitized solar cells (DSSC) based on liquid electrolyte and cyanoacrylate gel electrolyte were fabricated and devices *J/V* curves were measured. The effects of co-adsorbent chenodeoxycholic acid (CDCA), weight content of cyanoacrylate and cross linking polymer gel network on device efficiency were discussed. Under solar illumination of 100 mW cm<sup>-2</sup> (A.M. 1.5), the optimized liquid and quasi-solid-state devices efficiency of 8.21% and 5.93% were obtained, respectively. The long term stability is also presented.

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### 1. Introduction

Since the dye sensitized solar cell (DSSC) was firstly reported in 1991 by O'Regan and Grätzel [1], It has aroused intensive interest over the past two decades due to its low cost, simple preparation procedure [2,3] and validated efficiency of almost 13% [4]. In order to improve the photovoltaic performance of DSSC, extensive efforts have been focused on the sensitizers with high molar extinction coefficients and long term stable electrolytes [5,6]. Various kinds of ruthenium dye, cis-RuLL'(SCN)<sub>2</sub>, where L and L' are substituted bipyridyl ligands featuring different groups, with high molar extinction coefficients have still been synthesized and utilized in DSSC [7–10]. A new ruthenium sensitizer, Ru(2,2'-bipyridine-4,4'-dicarboxylic acid)-4,4'-bis(2-(2,4,6-tri-methoxyphenyl)ethenyl)-2,2'-bipyridine) (NCS)<sub>2</sub>, denoted K77-3 (Scheme 1) was synthesized and its photovoltaic performance was discussed in this paper.

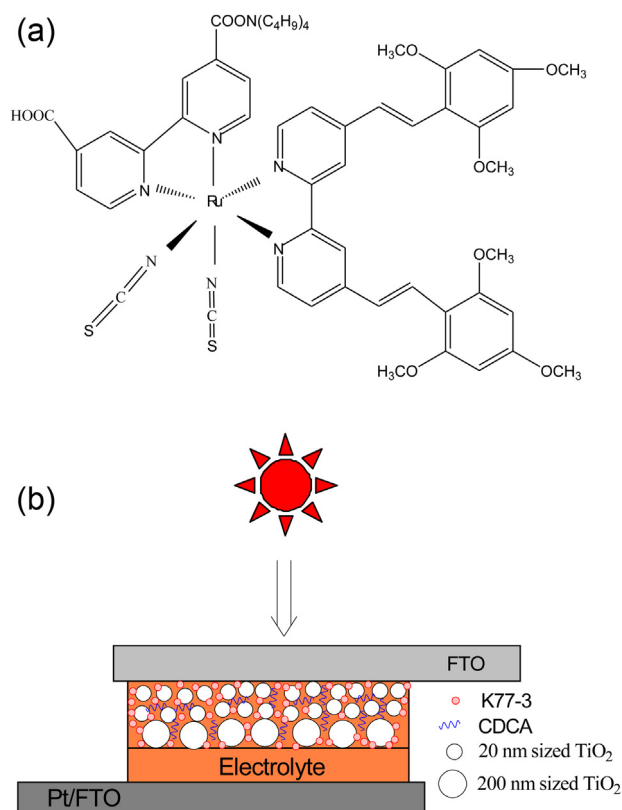
DSSCs based on liquid electrolytes have been achieved very high efficiency [4]. However the liquid electrolyte has some major technological disadvantages, such as the leakage and volatilization of liquid, which breaks down the long term performance and

limits practical use of the DSSC. Therefore large efforts have been made into replacing the liquid electrolyte by inorganic and organic semiconductors [11–14], solid polymer electrolytes [15–18] and molten hole conductor [19] [<http://www.sciencedirect.com/science/article/pii/S0379677911003742-cor0010-cor0010>]. However, due to low ionic conductivity, imperfect soakage of porous TiO<sub>2</sub> film, poor contact with counter electrode, and high recombination rate at the TiO<sub>2</sub>/solid-state-electrolyte interface, the energy conversion efficiency of DSSC based on the solid-state electrolytes were less than 5% [20].

Quasi-solid-state electrolyte, such as polymer gel electrolyte, can avoid the leakage and volatilization of liquid electrolyte and keep a high ionic conductivity and good interface contact with porous TiO<sub>2</sub> film and counter electrode, which is possible to accelerate the practical application of DSSC. There are two kinds of polymer gel electrolyte, one is non-reactive polymers, such as poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) [21], poly(methyl methacrylate)(PMMA) [22], poly(acrylic acid) (PAA) and poly(ethylene glycol) (PEG) hybrid [23], above 6% efficiency was achieved. The other polymer gel electrolyte used was reactive polymers which can form a three-dimensional molecular network as solid matrix in which the charge-carrying ions can move. High efficiencies have been reported from three such approaches using different chemistry to achieve a network [24–26]. Our group also reported one novel quasi-solid-state DSSC employing SuperGlue® (commercial glue) as electrolyte matrix [27]. Super glue has small size of cyanoacrylate monomers as well as their low viscosity, which allows an easy penetration into the TiO<sub>2</sub> pores. When

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**Scheme 1.** (a) Chemical structure of K77-3 and (b) DSSC device configuration.

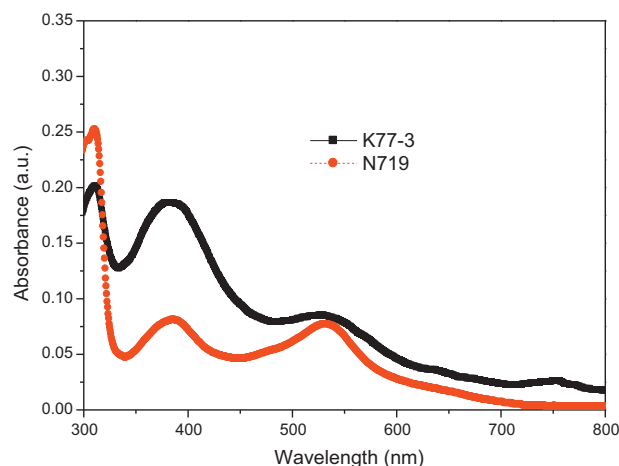
cianoacrylate was accounted with oxygen and water, it will be fast polymerized, which offers significant advantages in the fabrication of solar cells and modules as it is in itself a very good laminating agent. In this paper, one drop of cross linker of divinyltetramethylsiloxane was added in cyanoacrylate based gel electrolyte, the cross linking network will be formed, which will keep more liquid electrolyte component than pure cyanoacrylate polymer only bearing line type polymer structure. The influences of cross linker resulted in possible cyanoacrylate polymer network structure based on new dye K77-3 on DSSC device efficiency and long term stability are presented in this paper.

## 2. Experimental

### 2.1. Materials and synthesis

4,4'-Dimethyl-2,2'-bipyridine, 4,4'-dicarboxy-2,2'-bipyridine,  $\text{H}_2\text{SO}_4$ ,  $\text{CrO}_3$ , Potassium tert-butoxide,  $[\text{RuCl}_2(\text{p-cymene})]_2$ ,  $\text{NH}_4\text{NCS}$ , 2,4,6-trimethoxybenzaldehyde, chenodeoxycholic acid (CDCA), Titanium isopropoxide (TIP, 99.99%), guanidinium thiocyanate (GuNCS), 1,3-dimethylimidazolium iodide, iodine, 1-methylbenzimidazole and divinyltetramethylsiloxane used in this study are all reagent grade and used as purchased from Aldrich or Acros without further purification. Cyanoacrylate was purchased from supermarket as "SuperGlue<sup>®</sup>". Dimethylformamide (DMF) and 3-methoxypropionitrile were dried over molecular sieves (4 Å) for 24 h, then vacuum distilled under nitrogen atmosphere.

In the present work, a novel Ru complex sensitizer K77-3 (Scheme 1) was synthesized and its synthetic route was according to reference [5], the small modification was using 2,4,6-trimethoxybenzaldehyde to substitute 4-tert-butylbenzaldehyde. The detailed synthetic route and structure characterization will be published elsewhere.



**Fig. 1.** UV-vis absorption spectra of K77-3 and N719 in DMF and acetonitrile solution, respectively.

### 2.2. Device fabrication

FTO coated glass substrates were firstly cleaned by ultrasonication in acetone and isopropanol, and treated with 50 mM  $\text{TiCl}_4$  aqueous solution at 70 °C for 30 min in order to make a good mechanical contact between the following printed  $\text{TiO}_2$  layer and FTO glass electrode. Then a transparent film of 20 nm sized  $\text{TiO}_2$  particles was screen printed on the  $\text{TiCl}_4$ -treated FTO and further coated by the second layer of 200 nm sized light scattering anatase  $\text{TiO}_2$  particles. These two kinds of  $\text{TiO}_2$  paste were prepared according to Ref. [28]. The screen printed double-layer was heated to 500 °C under oxygen for 30 min and subsequently cooled to room temperature. After treatment with 50 mM  $\text{TiCl}_4$  aqueous solution again, the  $\text{TiO}_2$  films were rinsed with water and ethanol and then sintered at 500 °C for 30 min. The final thickness for the transparent layer is 10  $\mu\text{m}$  and that for the scattering layer is 4  $\mu\text{m}$  measured by the Surface profilometer (Tencor Alpha-500). After cooling to 80 °C, the titanium oxide electrodes were immersed into 0.3 mM solution of K77-3 dye or K77-3 and CDCA (0.3 mM: 0.3 mM) mixture in DMF in dry atmosphere at room temperature for 24 h.

The liquid electrolyte solution was prepared by dissolving 1,3-dimethylimidazolium iodide (1.0 M), iodine (0.15 M), 1-methylbenzimidazole (0.5 M), GuNCS (0.1 M) in 3-methoxypropionitrile. Quasi-solid-state electrolyte is prepared by adding some amount of cyanoacrylate into the above liquid electrolyte. One drop of electrolyte solution was put on the sensitized  $\text{TiO}_2$  electrode which was immediately pressed together with FTO on which a ca. 200 nm thick layer of Pt was thermally evaporated. To provide stability, the sides are covered with "SuperGlue<sup>®</sup>" and Epoxy adhesive. The active area of the cells was about 0.25  $\text{cm}^2$  determined by the cross section of Pt electrode and titanium oxide photoanode.

### 2.3. Characterization

UV-vis absorption spectra were recorded with Hitachi UV-Visible spectrophotometer (U-3000). The photocurrent density/voltage ( $J/V$ ) characteristics of the devices were measured using a Keithley 2400 under A.M. 1.5 (100  $\text{mW cm}^{-2}$ ) illumination.

## 3. Results and discussion

The UV-vis absorption spectrum of K77-3 dye dissolved in DMF is shown in Fig. 1. The absorption spectrum has a peak band corresponding to the lowest metal-to-ligand charge

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