



Mass transport effect on the photovoltaic performance of ruthenium-based quasi-solid dye sensitized solar cells using cobalt based redox couples



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ABSTRACT

Dye-sensitized solar cell (DSSC) technology is a promising solution to global energy and environmental problems. However, device stability and long-term durability are still significant issues in the commercial application of DSSC. Quasi-solid-state polymer electrolytes can be used in DSSCs in order to overcome various problems associated with liquid electrolytes. In this work we have studied the effect of mass transport on the photovoltaic performance of established ruthenium sensitizers (N719 and Z907) in DSSCs using the series of cobalt (II/III) quasi-solid electrolytes based on the poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) polymer. The $[\text{Co}(\text{bpy})_3]^{3+/2+}$ electrolyte showed better mass transport among all electrolyte systems examined. The DSSCs employing Z907 sensitizer exhibit significantly enhanced photovoltaic performances, in which a better power conversion efficiency of 4.34% was obtained from $[\text{Co}(\text{bpy})_3]^{3+/2+}$ redox mediator under 1 sun condition. The Z907 device efficiency significantly increased to 5.2% (2.66% under 1 sun) and sustained 100% photocurrent of the liquid electrolyte for $[\text{Co}(\text{dtb})_3]^{3+/2+}$ redox couple under 0.29 sun condition.

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1. Introduction

Over the past few years, tremendous research effort has been proffered to dye-sensitized solar cells (DSSCs) [1–6] due to their relatively low cost and high energy conversion efficiency since a power conversion efficiency (η) of higher than 10% was demonstrated for a DSSC first in 1993 by Grätzel et al., using *cis*-dithiocyanatobis (4,4'-dicarboxy-2,2'-bipyridine) ruthenium (II) (N3) as the sensitizer in conjunction with I^-/I_3^- redox electrolyte [7]. The I^-/I_3^- redox couple is also known to have limitations due to its large local concentration, complex redox chemistry, competitive light absorption and its corrosive nature, which effect the long term durability and large scale DSSCs production [8–13]. For outdoor application it is, however, of interest to find new redox mediator to I^-/I_3^- redox couple. Latterly, alternative redox mediators such as disulfide/thiolate [14], 2-mercapto-5-methyl-1,3,4-thiadiazole and its disulfide dimer [15] and tetramethyl formaminium disulfide/

tetramethylthiourea [16,17] have shown promising results compared with conventional I^-/I_3^- redox couple. However, these systems are similar in complexity to I^-/I_3^- system because they involve transfer of two electrons in their overall redox reactions.

In comparison to I^-/I_3^- , the one-electron $\text{Co}^{3+/2+}$ redox couples possess the advantage of non-corrosiveness and negligible visible light absorption [18–25]. Moreover, the electrochemical potential of these species can be adjusted through the modification of the ligand skeleton. This tuning minimizes energy losses in the cell by optimizing the driving force for regeneration, which allows for considerable improvement of the open-circuit photovoltage relative to I^-/I_3^- electrolytes. Recently, porphyrin-sensitized devices with a $\text{Co}^{3+/2+}$ redox electrolyte showed an efficiency of 13% [25]. However, the ultimate goal of DSSC research is to facilitate the commercialization and large scale deployment of DSSCs. The presence of organic solvents, such as acetonitrile and 3-methoxypropionitrile (MPN), in DSSCs can result in some practical limitations sealing and long-term operation. Gelation of the liquid electrolytes mitigate the potential instability against solvent leakage under thermal stress. DSSCs based on gel electrolyte can compete with liquid DSSCs in terms of power conversion efficiencies and, importantly, they have been reported to show better

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thermal stability [26]. For example, poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) based MPN (3-methoxypropionitrile) quasi-solid state DSSCs have been reported with efficiencies over 6% and good long term stability [27]. To the best of our knowledge, 9.5% is the highest efficiency that has been reported for a polymer gel based DSSC at 1 sun illumination using I^-/I_3^- redox mediator [28]. To date, however, there are only two publications reporting the application of cobalt-based gel electrolytes in DSSCs [29]. In this context, cobalt-based redox mediators incorporating polymer or gel materials were introduced to substitute the I^-/I_3^- redox system. The profitable application of PVDF-HFP in DSSC polymer gel electrolytes containing the $Co^{3+/2+}$ redox shuttle, directed us to investigate the series of cobalt based polymer gel electrolytes. Finally, we have characterized the photovoltaic performance of DSSCs containing the well-known standard and stable sensitizers N719 and Z907 in the presence of I^-/I_3^- and three different $Co^{3+/2+}$ polymer gel mediators. The chemical structures of ruthenium sensitizer N719, Z907 and polymer PVDF-HFP are shown in Fig. 1. The redox species were cobalt(III/II) tris(4,4'-di-*tert*-butyl-2,2'-bipyridine), $[Co(dtb)_3]^{3+/2+}$, cobalt(III/II) tris(2,2'-bipyridine), $[Co(bpy)_3]^{3+/2+}$ and cobalt(III/II) tris(1,10-phenanthroline), $[Co(phen)_3]^{3+/2+}$. An energy level diagram of a DSSC sensitized with N719 and Z907, employing the different cobalt redox couples and compared here, and the chemical structures of the complexes are shown in Fig. 2a and b, respectively. The DSSC sensitized with Z907 using $[Co(bpy)_3]^{3+/2+}$ redox couple exhibited an efficiency of 4.34% under 1 sun which reached the 70% of the iodine based quasi-solid electrolyte. Moreover, we demonstrate that the mass transport limitations are also efficiently avoided at low light intensities and at 0.29 sun the Z907 sensitizer reached an efficiency of over 5.2% for $[Co(dtb)_3]^{3+/2+}$ redox couple.

2. Experimental

2.1. Materials and measurements

The cobalt complexes $[Co(bpy)_3](PF_6)_2$, $[Co(bpy)_3](PF_6)_3$ (*bpy* = 2,2'-bipyridine), $[Co(phen)_3](PF_6)_2$, $[Co(phen)_3](PF_6)_3$ (*phen* = 1,10-phenanthroline), $[Co(dtb)_3](PF_6)_2$ and $[Co(dtb)_3](PF_6)_3$ (*dtb* = 4,4'-di-*tert*-butyl-2,2'-dipyridyl) were synthesized according to a previously reported procedure. Lithium perchlorate ($LiClO_4$), 4-*tert*-butylpyridine (*tBP*), iodine (I_2), 1,2-dimethyl-3-propylimidazolium iodide (*PMII*), 1-butyl-3-methylimidazolium iodide (*BMI*) and solvents were used as received from Sigma-Aldrich and TCI.

Absorption spectra were recorded in a quartz cell with 1 cm path length on a UV–Vis spectrometer (HP8453, PDA type). 1H NMR spectra were recorded on a Varian Mercury NMR 300 MHz

spectrometer. The reported chemical shifts are relative to TMS. The photocurrent-voltage measurement was performed using a Keithley model 2400 Source Meter and a Newport 91192 solar simulator system (equipped with a 1 kW xenon arc lamp, Oriel). Light intensity was adjusted to 1 sun (100 mW/cm^2) with a Radiant Power Energy Meter (model 70260, Oriel). The incident photon-to-current conversion efficiency (IPCE) results were acquired from IPCE G1218a (PV Measurement). This system applies monochromatic light from a 75 W xenon arc lamp (Ushio UXL-75XE) filtered by a dual-grating monochromator and individual filters onto the test devices. An ellipsoidal reflector collects light from the lamp and focuses on the monochromatic entrance slit via a mechanical chopper to create a small modulated signal. While the modulated, monochromatic light was applied to the test devices, a continuous bias light (ca. 1 sun) was also applied. Electrochemical impedance spectroscopy (EIS) was performed using an electronic-chemical analyzer (Iviumstat Tec).

2.2. Synthesis of $[Co(bpy)_3](PF_6)_2$ and $[Co(bpy)_3](PF_6)_3$ complexes

2,2'-Bipyridine (2.450 g, 15.69 mmol) was dissolved in a minimum of methanol. $CoCl_2 \cdot 6H_2O$ (1.164 g, 4.89 mmol) was then added drop wise while stirring. After stirring for about 30 min, potassium hexafluorophosphate (2.700 g, 14.67 mmol) in distilled water was added and then stirring for 2 h. The precipitated complex was filtered, washed with distilled water and methanol, and dried in an oven to obtain $[Co(bpy)_3](PF_6)_2$ (4.000 g).

$[Co(bpy)_3](PF_6)_2$ ($C_{30}H_{24}CoF_{12}N_6P_2$) Yield 90%, 1H NMR (300 MHz, $DMSO-d_6$): δ 8.710 (d, $J = 3.3$ Hz, 6H), 8.413 (d, $J = 7.5$ Hz, 6H), 7.989 (t, $J = 8.1$ Hz, 6H), 7.474 (t, $J = 6.1$ Hz, 6H). In the ^{13}C NMR spectrum 5 signals were found at δ (ppm) = 155.1, 149.1, 137.1, 124.0 and 120.2. IR (KBr): ν_{max}/cm^{-1} : 1569, 1600, 2351 and 3109. Elemental analysis calculated for $C_{30}H_{24}CoF_{12}N_6P_2$: C, 44.08; H, 2.96; N, 10.28%. Found: C, 44.02; H, 2.97; N, 10.65%.

2,2'-Bipyridine (1.560 g, 9.98 mmol) was dissolved in a minimum of methanol. $CoCl_2 \cdot 6H_2O$ (0.742 g, 3.12 mmol) was then added drop wise while stirring. Hydrogen peroxide (7.000 g) and hydrochloric acid (3.100 g) were then added in equivalent molar concentration, and stirring continued for about 1 h. Potassium hexafluorophosphate (2.300 g, 12.50 mmol) in distilled water was added to form a precipitate. The precipitated complex was filtered, washed with de-ionized water and methanol, and dried in an oven to obtain $[Co(bpy)_3](PF_6)_3$ (3.000 g).

$[Co(bpy)_3](PF_6)_3$ ($C_{30}H_{24}CoF_{18}N_6P_3$) Yield 60%, 1H NMR (300 MHz, $DMSO-d_6$): δ 9.031 (d, $J = 7.2$, 6H), 8.583 (t, $J = 7.5$ Hz, 6H), 7.786 (t, $J = 7.0$ Hz, 6H), 7.428 (d, $J = 5.4$ Hz, 6H). In the ^{13}C NMR spectrum 5 signals were found at δ (ppm) = 155.1, 149.1, 137.1, 124.0 and 120.2. IR (KBr): ν_{max}/cm^{-1} : 1506, 1606, 2364 and 3128. Elemental analysis

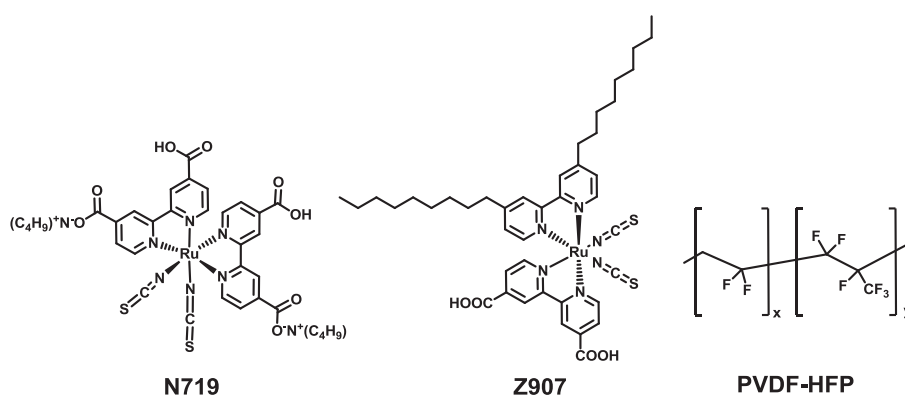


Fig. 1. Chemical structures of ruthenium sensitizers (N719 and Z907) and polymer (PVDF-HFP).

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