

Influence of alkyl dihalide gelators on solidification of dye-sensitized solar cells

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Abstract

Quasi-dye-sensitized solar cells were prepared by using ionic liquid-type electrolytes and gelators consisting of polyvinylpyridine and alkyl dihalides. Gelation occurred by the reaction of polyvinylpyridine and alkyl dihalides. When the chain length of the dihalides was varied, the short-circuit current (J_{sc}) increased with an increase in the chain length. However, the open-circuit voltage (V_{oc}) and fill factor (ff) slightly decreased. The increase in J_{sc} was brought about by the decrease in the interfacial resistances between the gel electrolyte and the counter electrode. In addition, the increase in the J_{sc} was explained by increases in the apparent diffusion coefficient of I^-/I_3^- when the chain length increased. Decreases in V_{oc} and ff were explained by back-electron transfers from TiO_2 to iodine in the electrolytes. V_{oc} of the cells solidified by alkyldiiodide was lower than that solidified by alkyldichloride or alkyldibromide. It was explained by negatively shifted redox potential of I^-/I_3^- , compared with those for Cl^-/Cl_2 or Br^-/Br_2 .

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1. Introduction

Dye-sensitized solar cells (DSCs) are considered to be promising candidates for conventional silicon solar cells, because of the comparatively high conversion efficiency, up to 10%, and their low production cost [1]. One of the drawbacks of DSCs for practical use is that they consist of a volatile organic solvent. Quasi-solidification of the electrolyte has been reported to overcome this problem [2–9]. However, the problems on solvent evaporation still remain. We reported quasi-solid dye-sensitized solar cells consisting of chemically cross-linked ionic liquid-gel electrolytes [10–17]. The ionic liquid electrolytes were solidified by the reaction of polyvinylpyridine with multi-functional halides acting as cross-linkers. The ionic liquids decreased their volatility and the gels prevented the liquid electrolytes from leaking. Tetrakis(bromomethyl)benzene was found to be an excellent cross-linker, which caused

phase-separated gel electrolytes [13]. To shed the light on the fundamental aspect on gel electrolytes, simple alkyl dihalides as the cross-linkers of polyvinylpyridine were used. In this paper, we focus on the relationship between the alkyl dihalides and the photovoltaic performances, and clarify the role of the alkyl dihalides.

2. Experimental

cis-Bis(isothiocyanato)-*N,N'*-bis(2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium (referred to as N3 dye) and TiO_2 paste (Ti-Nanoxide D) were purchased from Solaronix SA. Fluorine-doped SnO_2 layered glass plates ($7\text{--}8\ \Omega$ square⁻¹)(SnO_2/F) were obtained from the Nippon Sheet Glass Co. Ltd., poly(4-vinylpyridine)(PVP)(MW:60,000), 1,2,4,5-tetrakis(bromomethyl)benzene(B4Br), and alkyldi-halides were purchased from the Aldrich Corporation.

Chemical structures and abbreviations of alkyl halides as cross-linking agents are shown in Fig. 1. All the reagents were commercially available and were used without further purification. The carbon length of the alkyl groups and the halide moieties were varied. B4Br was also employed as a

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reference cross-linker. Table 1 shows compositions of gel electrolyte precursors. Gel electrolyte precursors consisted of poly(4-vinylpyridine)(PVP) and the equivalent ratio of the cross-linking agent (alkyl dihalide) in ionic liquid electrolyte (IL). IL consisted of I_2 (300 mM) in 3-methyl-1-propylimidazolium iodide (MePrImI) containing 5% water [10–17]. LiI was not added into the electrolytes to make the electrolyte composition simple, which enables one to compare the characteristics of the alkyl dihalide cross-linkers clearly.

Ti–Nanoxide D paste was coated on SnO_2/F layered glasses by using a metal mask (t: 30 μm) and baked at 450 °C for 30 min. The TiO_2 layers were immersed in N3 dye solutions in ethanol (0.1%) at room temperature overnight. After these TiO_2 electrodes were rinsed with dry ethanol, they were dried at room temperature in the dark. Pt-sputtered SnO_2/F glass substrates were used as the counter electrodes. A plastic sheet (Himiran, Mitsui-Dupont Co. Ltd.) was inserted between the TiO_2 electrodes and the counter electrodes as a spacer. Gel electrolyte precursors were injected in the cell at room temperature. Then the cells were heated at 80 °C for 5 min to form gels.

The cross-linking was carried out by the reaction of PVP with alkyl dihalides by addition reactions as shown in Scheme 1. Finally, the cells were encapsulated with epoxy resins. The cell area was 1 cm^2 .

Charge-transfer resistances were monitored by the method described in the previous literature [18]. The symmetrical cells, which were sandwiched with two sputtered Pt electrodes fabricated on the SnO_2/F layered glass substrates, were measured. The charge-transfer resistances between Pt electrodes and gel electrolytes were estimated by using a Cole–Cole plot [18]. Impedance spectra were recorded over frequency ranges from 0.01 Hz to 100 kHz using a Solartron model 1255B frequency response analyzer. Photo-electrochemical measurements were performed using an AM1.5 solar simulator (WXS-R50S-1.5, Maki Manufacturing) furnished with a xenon lamp. The light intensity was monitored with a polysilicon illuminometer standardized by the Japan Quality Assurance Organization (JQA). Since the photovoltaic performance depended on how to make cells and their conditions, the performance of various cells were compared with each other in the same series of experiments.

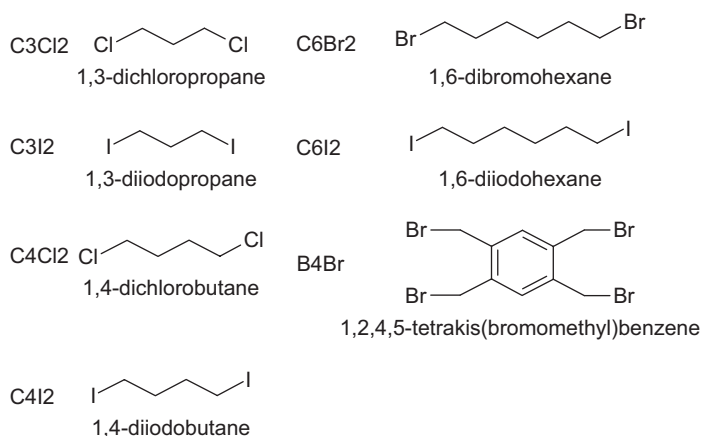


Fig. 1. Abbreviation for various cross-linking agents tested in this work.

Table 1
Compositions for gel electrolyte precursors

Abbreviation	Gelator		Cross-linker		Electrolytes parts
	Backbone oligomer				
	Structure	Wt%	Structure	Wt% (mmol%)	
GP1	PVP	4	C3Cl2	2.2 (19.5)	100
GP2	PVP	4	C3I2	5.6 (18.9)	100
GP3	PVP	4	C4Cl2	2.4 (18.9)	100
GP4	PVP	4	C4I2	5.9 (19.0)	100
GP5	PVP	4	C6Br2	4.6 (18.9)	100
GP6	PVP	4	C6I2	6.4 (18.9)	100
GP7	PVP	2	B4Br	2.0 (4.4)	100

Electrolyte: 1-methyl-3-propylimidazolium iodide (MePrImI) containing 5 wt% of water, I_2 : 300 mM, PVP: polyvinylpyridine; C3Cl2: 1,3-dichloropropane; C3I2: 1,3-diiodopropane; C4Cl2: 1,4-dichlorobutane; C4I2: 1,4-diiodobutane; C6Br2: 1,6-dibromohexane; C6I2: 1,6-diiodohexane; B4Br: 1,2,4,5-tetrakis(bromomethyl)benzene.

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