

Synthesis of Thiocyanate Molten Salts and its Application to the Solid-State Dye-Sensitized Photovoltaic Cells

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Synthesis of thiocyanate molten salts and its application to the solid-state dye-sensitized photovoltaic cells Akinori Konno,[†] G. R. Asoka Kumara,[†] and Kirthi Tennakone[‡] [†]Faculty of Engineering, Shizuoka University, 3-5-1 Johoku, Hamamatsu, 432-8561 Japan. [‡]Institute of Fundamental Studies, Hantana road, Kandy, Sri Lanka.

Room temperature molten salts constitute promising systems with the potential of practical applications in a diverse variety of technological disciplines.¹ Organic electrochemical applications of these materials have intensively been studied especially for environmentally friendly alternative to the conventional organic electrolytic medium, i.e. supporting electrolyte and organic solvent. An interesting observation we made was the peculiar behavior of the thiocyanate ion. Some alkylammonium thiocyanates were also reported to show quite low melting points.² On the other hand, we have reported successful application of one of these 1-ethyl-3-methylimidazolium thiocyanates, i.e. thiocyanate, to the dye-sensitized solid-state solar cell, as an additive and a crystal growth inhibitor for CuI layer preparation.³ The preparation of novel room temperature molten thiocyanate salts and their preliminary application to the solid-state dye-sensitized photovoltaic cells (DSSSCs) were also reported previously.⁴ The structure of DSSSC is shown in figure 1. In this paper, development of novel room temperature molten thiocyanate salts and their effect on the performance of DSSSCs are presented in detail.

Several kinds of thiocyanates were prepared by the reaction of an alkyl amine with NH₄SCN. It is noted that this reaction was greatly promoted by microwave irradiation and completed within 10 min. All of them resulted in the formation of room temperature molten thiocyanates (Table 1). The alkylammonium thiocyanate which have a hydroxyl group in alkyl chain (DMAE-SCN and DEAE-SCN) showed lower melting point. On the other hand, the conductivity of them were lower than that of TEA-SCN. In contrast to these ammonium thiocyanates posessing hydroxyl group, tris(2hydroxyethyl)ammonium thiocyanate (NTE-SCN) showed higher melting point and lower conductivity.

We investigated the effect of these thiocyanates as an additive in CuI layer of DSSSC. The performances of TiO₂/Dye/CuI cell with the addition of a variety of thiocyanate (4.5 mol%) to CuI layer, are shown in table 2. In the cases of DMAE and DEAE-SCN, the addition of these molten thiocyanates resulted in lower performance of TiO₂/Dye/CuI DSSSCs than the case of TEA-SCN mainly because of lower photocurrent density (Jsc). It is reasonable considering lower conductivity of DMAE and DEAE-SCN than that of TEA-SCN. On the other hand, it is remarkable that the addition of NTE-SCN to CuI layer resulted in higher photocurrent than TEA-SCN.

References

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Figure 1. Schematic structure of TiO₂/Dye/CuI cell.

Table 1. Properties of alkylammonium thiocyanates.



 $\begin{array}{ll} {\sf TEA-SCN} & : \ {\sf R}^1 {\sf =} \ {\sf R}^2 {\sf =} \ {\sf R}^3 {\sf =} \ {\sf CH}_2 {\sf CH}_3 \\ \\ {\sf DMAE-SCN} & : \ {\sf R}^1 {\sf =} \ {\sf R}^2 {\sf =} \ {\sf CH}_3, \ {\sf R}^3 {\sf =} \ {\sf CH}_2 {\sf CH}_2 {\sf OH} \\ \\ {\sf DEAE-SCN} & : \ {\sf R}^1 {\sf =} \ {\sf R}^2 {\sf =} \ {\sf CH}_2 {\sf CH}_3, \ {\sf R}^3 {\sf =} \ {\sf CH}_2 {\sf CH}_2 {\sf OH} \\ \\ {\sf NTE-SCN} & : \ {\sf R}^1 {\sf =} \ {\sf R}^2 {\sf =} \ {\sf R}^3 {\sf =} \ {\sf CH}_2 {\sf CH}_2 {\sf OH} \\ \end{array}$

Thiocyanate	m.p./°C	Conductivity ^a $\log (\sigma / \text{Scm}^{-1})$	
TEA-SCN	15	-2.21	
DMAE-SCN	-102	-2.80	
DEAE-SCN	-100	-2.63	
NTE-SCN	29	-3.01	

^a Measured at 25 °C.

Table 2. Solar cell performance of TiO₂/N3 dye/CuI DSSSCs with the addition of a variety of thiocyanate (4.5 mol%) to CuI layer

thiocyanate	Voc / V	Jsc / mAcm ⁻²	Fill Factor	Efficiency /%
TEA-SCN	0.41	8.2	0.55	1.9
DMAE-SCN	0.38	7.6	0.53	1.5
DEAE-SCN	0.43	6.8	0.52	1.5
NTE-SCN	0.35	9.0	0.54	1.7