

## On the addition of conducting ceramic nanoparticles in solvent-free ionic liquid electrolyte for dye-sensitized solar cells<sup>☆</sup>

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### ABSTRACT

Titanium carbide (TiC) is an extremely hard conducting ceramic material often used as a coating for titanium alloys as well as steel and aluminum components to improve their surface properties. In this study, conducting ceramic nanoparticles (CCNPs) have been used, for the first time, in dye-sensitized solar cells (DSSCs), and the incorporation of TiC nanoparticles in a binary ionic liquid electrolyte on the cell performance has been investigated.

Cell conversion efficiency with 0.6 wt% TiC reached 1.68%, which was higher than that without adding TiC (1.18%); however, cell efficiency decreased when the TiC content reached 1.0 wt%. The electrochemical impedance spectroscopy (EIS) technique was employed to analyze the interfacial resistance in DSSCs, and it was found that the resistance of the charge-transfer process at the Pt counter electrode ( $R_{ct1}$ ) decreased when up to 1.0 wt% TiC was added. Presumably, this was due to the formation of the extended electron transfer surface (EETS) which facilitates electron transfer to the bulk electrolyte, resulting in a decrease of the dark current, whereby the open-circuit potential ( $V_{OC}$ ) could be improved. Furthermore, a significant increase in the fill factor (FF) for all TiC additions was related to the decrease in the series resistance ( $R_s$ ) of the DSSCs. However, at 1.0 wt% TiC, the largest charge-transfer resistance at the  $TiO_2$ /dye/electrolyte interface was observed and resulted from the poor penetration of the electrolyte into the porous  $TiO_2$ . The long-term stability of DSSCs with a binary ionic liquid electrolyte, which is superior to that of an organic solvent-based electrolyte, was also studied.

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### 1. Introduction

The rising price of fossil fuels, together with their rapid depletion and the pollution caused by their combustion, is forcing us to find sources of clean renewable energy. Solar power is considered to be one of the best sustainable energies for future generations. Recently, dye-sensitized solar cells (DSSCs) have attracted much attention as a possibly highly efficient and low-cost alternative to conventional silicon solar cells [1]. A DSSC is composed of three adjacent thin layers: a mesoporous oxide film, such as  $TiO_2$  supported on transparent conducting glass; dye molecules, such as ruthenium bipyridyl derivatives which are sensitive to visible light in the solar spectrum, and an electrolyte

solution containing iodide and triiodide ions as a redox mediator to reduce the oxidized dye molecules. The three layers are sandwiched together by a second conducting glass covered with platinum [2].

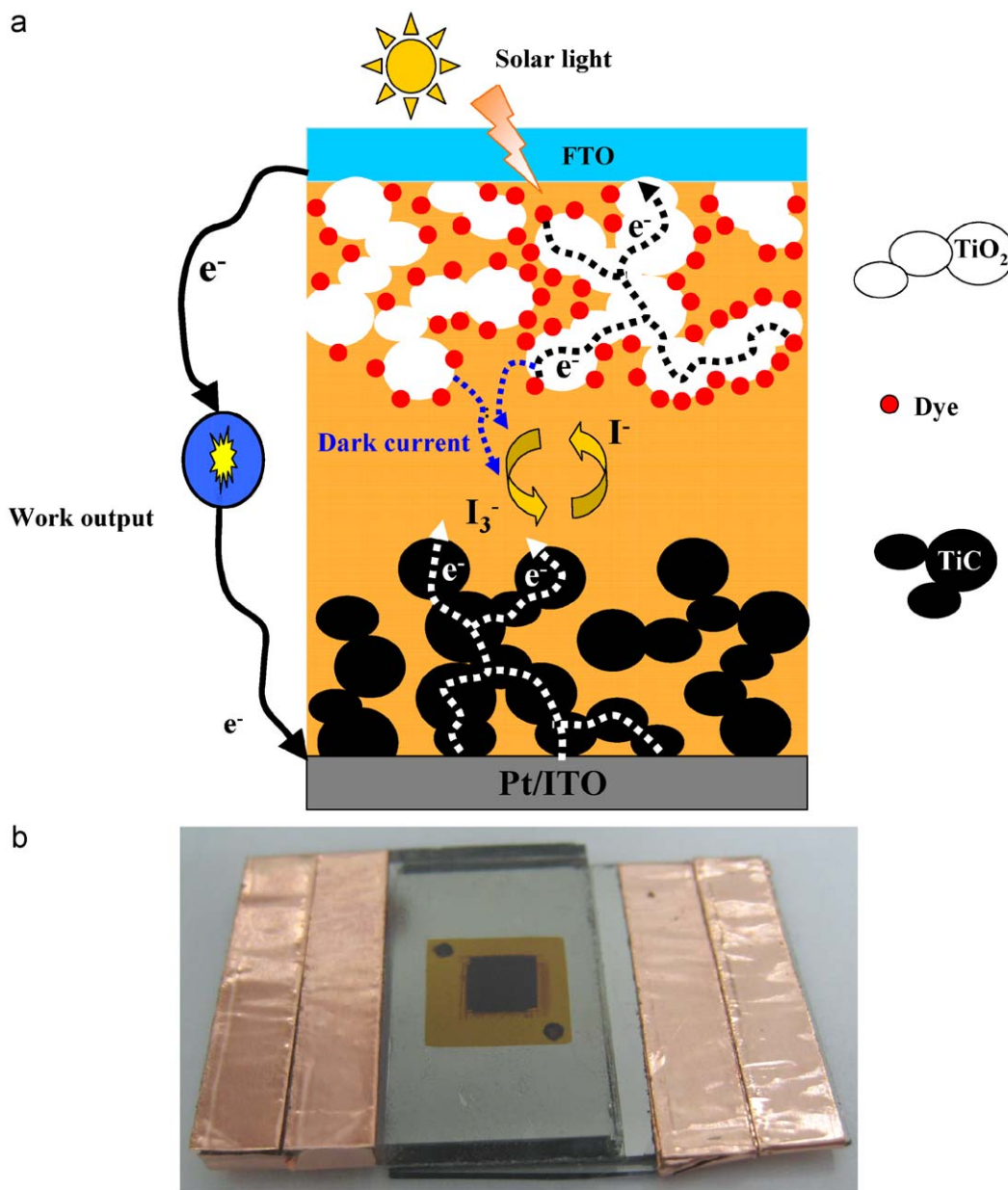
However, leakage and the volatilization of organic solvent-based electrolytes have restricted practical applications of DSSCs. Room-temperature ionic liquids (RTILs) have become attractive candidates for replacing organic solvents because of their negligible vapor pressure, high thermal stability, wide electrochemical window, and high ionic conductivity [3–7]. Nevertheless, the viscosity of RTILs is still much higher than that of organic solvents, including acetonitrile (ACN) and 3-methoxypropionitrile (MPN), and has resulted in lower power conversion efficiencies because of the RTILs' poor ionic diffusion ability.

TiC is an extremely hard conducting ceramic material, which is often used as a coating to improve the surface properties of metal surfaces [8]. In this study, titanium carbide (TiC) was added to ionic liquid-based electrolyte to form the extended electron transfer surface (EETS) (Scheme 1) from the counter electrode's surface to the bulk electrolyte, in order to facilitate electron transfer and, thereby, decrease the dark current from the working

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**Scheme 1.** (a) Schematic and (b) photograph of the actual DSSC device.

electrode to the electrolyte. The literature on the use of TiC has been limited to the surface coating of stainless steel, the Si/TiC composite anode for Li-ion batteries [9], and the TiC/C transition layers on coated graphite as thermal stress-relieving layers [10].

For the first time, TiC has been used in DSSCs, and the effect of TiC addition in the solvent-free ionic liquid electrolyte was studied.

## 2. Experimental section

Lithium iodide (LiI), iodine ( $I_2$ ), poly(ethylene glycol) (PEG, M.W. = 20,000), and 1-butyl-3-methylimidazolium iodide (BMII) were obtained from Merck; 4-tert-butylpyridine (TBP) and tert-butyl alcohol were obtained from Acros; and guanidine thiocyanate (GuSCN), 1-ethyl-3-methylimidazolium thiocyanate (EMISCN), titanium(IV) isopropoxide (TTIP), acetonitrile, acetylacetone, ethanol, neutral cleaner, and isopropyl alcohol (IPA) were obtained from Aldrich. Various amounts of titanium carbide

(Aldrich, 30 nm sized) were added to the binary ionic liquid-based electrolyte, composed of 0.6 M BMII, 0.2 M  $I_2$ , 0.5 M TBP, and 0.1 M GuSCN in EMISCN, to study the effect of TiC on the performance of DSSCs. For the organic solvent electrolyte, a mixture of 0.1 M LiI, 0.6 M DMPII (Solaronix S.A., Aubonne, Switzerland), and 0.05 M  $I_2$ , 0.5 M TBP in 3-methoxypropionitrile (MPN, Fluka) was used.

The commercial titanium dioxide (ST-21, 50 m<sup>2</sup>/g, 6.0 g, Ya Chung Industrial Co. Ltd., Taiwan) was thoroughly mixed with a solution of acetylacetone (500  $\mu$ l) in DI-water (11.0 g). This was stirred for 3 days and 1.8 g of PEG was then added to the well-dispersed colloid solution. The final mixture was stirred for an additional 2 days, and the TiO<sub>2</sub> paste was prepared.

The fluorine-doped SnO<sub>2</sub> conducting glass (FTO, 15  $\Omega$ /sq, Solaronix S.A., Aubonne, Switzerland) was first cleaned with a neutral cleaner, and then washed with DI-water, acetone, and IPA, sequentially. The conducting surface of the FTO was treated with a solution of TTIP (0.028 g) in ethanol (10 ml) in order to make a good mechanical contact between the following bladed TiO<sub>2</sub> layer

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