

CNT-Polymer composite 겔-전해질을 이용한 염료감응 태양전지에 관한 연구

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**Carbon nanotubes (CNTs)-polymer composite gel electrolyte
for solid-state dye sensitized solar cells (DSSCs)**

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Introduction

Owing to its simple structure with relatively high conversion efficiency (10%), dye-sensitized solar cells (DSSCs) are one of the promising candidates for the next generation solar cells, in contrast to inexpensive fabrication procedures of amorphous silicon [1-2]. Although DSSCs based on liquid electrolytes have reached efficiency as high as 10% under AM 1.5, the use of liquid electrolytes have created a lot of difficulties in sealing and long-term photochemical stability of the device [3-4]. Solid state and quasi-solid state electrolytes such as polymer composite, inorganic-inorganic composite materials have been used as promising materials to replace liquid electrolytes [5-6].

Recently, polymer nanocomposite electrolytes have known better thermal, mechanical and ionic conductivity as compared to conventional polymer composite electrolytes due to stronger interaction between polymer and nanofiller phases [7]. Recently, CNTs-polymer composite have shown the better interaction between filler and polymer which are responsible for the enhancement of ion conductivity. Nanocomposite electrolytes are a promising candidate to overcome the leakage and evaporation of liquid electrolyte in dye sensitized solar cells [8-9]. In this work, we described the preparation of CNTs-polymer composite electrolytes and fabricated the solid-state DSSCs. The fabricated DSSCs with 1% CNT-PEO electrolyte achieved moderately high conversion efficiency of 3.48% with an open circuit voltage (V_{oc}) of 0.589 volt and short circuit current (I_{sc}) of 10.64 mA/cm². The highly flexible nature of CNTs could be improved the interaction and cross-linking between nanofiller and polymer molecules which are significantly enhanced the electrical and ion conductivity properties of composite electrolytes.

Experimental

Commercial titanium dioxide powder (P25, Degussa) was used as TiO₂ sources. Ruthenium 535 bis-TBA dye (Solaronix) as a sensitizer and other chemicals were analytical grade and used as received. The transparent conducting oxide (TCO) glass substrate was supplied by

Hartfordglass company Inc. (TEC-8, FTO-coated glass, 8 /sq, 80% transmittance in the visible light).

Preparation of TiO₂ electrodes

For the preparation of TiO₂ thin-film, TiO₂ slurry was prepared by the incremental addition of 2 ml of aqueous polyethylene glycol (Fluka, average MW of 20,000) solution to 0.5 g of TiO₂ powder in a mortar under vigorous grinding with pestle. Each 0.1 ml of the aqueous polyethylene glycol solution was added after the previous mixture had formed a uniform and lump-free paste. Thus prepared uniform slurry was coated on TCO glass by a doctor blade technique. After natural drying at room temperature, the thin film was calcined in static air at 450°C for 30 min.

Preparation of gel electrolyte

The composite electrolytes were prepared by using 10% polyethylene oxide (PEO) solution in acetonitrile and different concentration of carbon nanotubes (CNTs) with LiI 0.1M, and I₂ 0.015M. The whole mixture was placed for the sonication to disperse all CNTs into the matrix of polymer. Lastly, the mixture kept on magnetic stirrer and stirred over a period of 24 hours for complete mixing between CNTs to polymer molecules.

Fabrication the Cells

To fabricate the DSSCs, thus prepared TiO₂ thin film electrodes were immersed in the dye solution of 0.3 mM ruthenium 535 bis-TBA in dry ethanol at room temperature for 24 hrs. The dye-adsorbed electrodes were then rinsed with ethanol and dried under a nitrogen stream. Pt counter electrodes were prepared by electron beam deposition of Pt (60 nm thickness) on ITO glass. The resulting dye adsorbed film was sealed with a Pt-sputtered conducting glass by a spacer (surlyn) and the gel electrolyte was introduced into the cell through one of two small holes drilled in the counter electrode.

The prepared sample was characterized by FE-SEM, (Hitachi 4700), AFM, XPS, FT-IR, DSC (TA analyzer), and ionic conductivity (digital conductometer). The photochemical characterization of DSSCs including photocurrent density was measured by using a scanning potentiostat (EG&G 273). The device was connected in a two-electrode configuration: the dye adsorbed TiO₂ film on TCO glass was connected as the working electrode and the Pt-coated TCO glass was used as the pseudo-reference (circuited with the counter electrode). Photocurrent–Voltage (I–V) curve was measured by using two computerized digital multimeters (Model 2000, Keithley) and a variable load. The light source was a 1000-W halogen lamp (Philips lighting) and its radiant power was adjusted with respect to Si reference solar cell (Fraunhofer Institute for Solar Energy System; Mono-Si + KG filter) to about one-sun light intensity (100 mW/cm²).

Results and discussion

FE-SEM of CNTs-polymer composite materials

FE-SEM of CNTs and CNTs-polymer composite are shown in Fig. 1a and b, respectively. CNTs have exhibits long tubular structures of 500-700nm with diameter of 10-15nm and all nanotubes are clear visible and separate. Fig.1b displayed the nanotubes are fully covered by the polymer molecules, it is indicating that the attachment and dispersion of CNTs on the surface of polymer. Therefore, the high penetration of polymer to CNTs may leads the high crosslinking and ion conductivity.

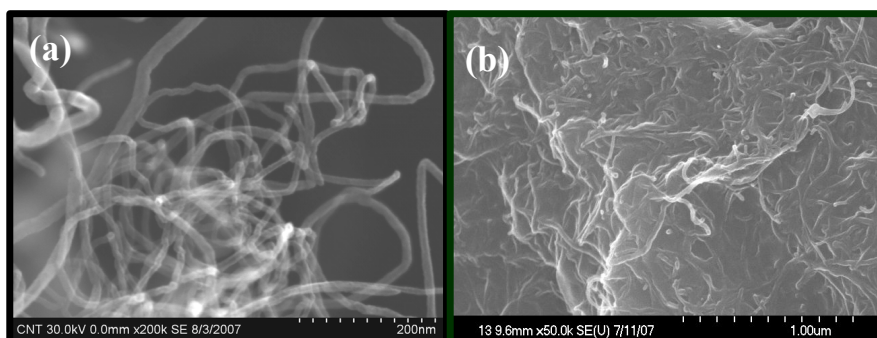


Figure-1 FE-SEM images of (a) CNTs and CNTs-Polymer composites

AFM of CNTs-polymer composite materials

Fig. 2a illustrates the surface of CNTs film obtained by the AFM in tapping mode. It shows well disperse and elongate tubular structure with length of 500-700nm and diameter of 10-15nm. Fig.2b shows the surface AFM image of CNTs-polymer composite. The surface of CNTs is fully and uniformly covered by the polymer molecules in composite materials. It is revealed that the size and length of CNTs increased in comparison with the original size because of the influence of polymer attachment to the wall of CNTs. This result is also fully supported the FE-SEM images of CNTs-polymer composite. Hence, the introduction of CNTs into the polymer matrix may cause the lowering of crystallinity of polymer, in excellent agreement with DSC results.

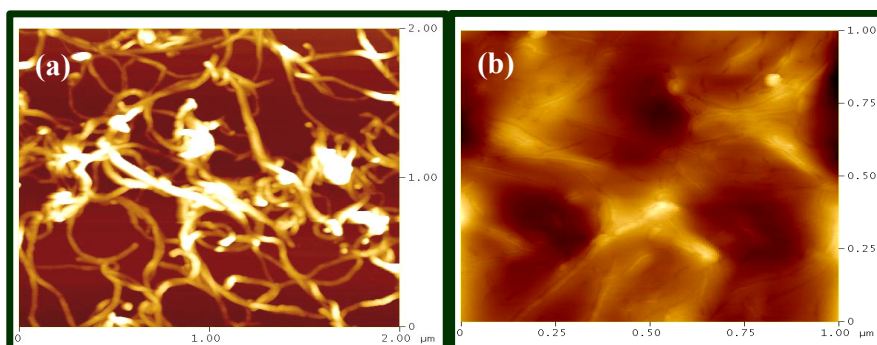
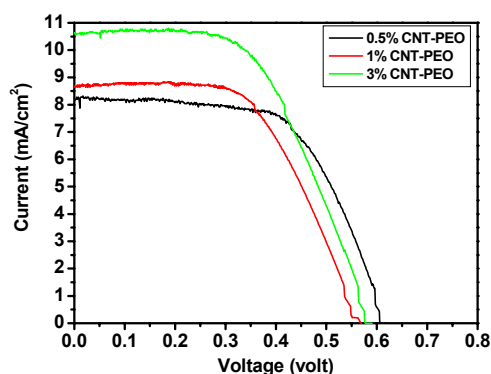


Figure-2 AFM images of (a) CNTs and CNTs-Polymer composites

Photovoltaic properties of CNTs-polymer electrolytes based DSSCs

The current (I) – voltage (V) curves of the solid-state DSSCs with CNTs-polymer composite electrolytes is illustrated in Fig.3. The Photovoltaic properties such as short circuit photocurrent density (J_{sc}), open circuit voltage (V_{oc}), fill factor (FF) and conversion efficiency (η) obtained are summarized in Table 1. The highest J_{sc} of 10.64 mA/cm^2 is observed in the DSSCs with CNTs (1%)-PEO electrolyte resulting in the conversion efficiency of 3.48 % which is higher than that with other compositions. The improvement in the performance of DSSCs is comparatively well in compare to the other solid-state DSSCs. The increase of J_{sc} , V_{oc} and conversion efficiency is rationalized in terms of strong interaction and bonding between CNTs and polymer molecules in composite electrolytes, which suppresses the surface recombination and improves the contact between the surface of electrolyte to TiO_2 electrode.

Table 1 Summary of IV-curves of DSSCs



Samples	J_{sc} (mA/cm^2)	V_{oc} (Volt)	FF (%)	η (%)
0.5%CNT-PEO	8.16	0.618	69.7	3.12
1%CNT-PEO	10.64	0.589	55.9	3.48
3%CNT-PEO	8.44	0.594	55.3	2.78

Figure 3 IV-Curves of DSSCs with CNTs-PEO Electrolytes

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