



# Performance enhancement of poly (vinylidene fluoride-co-hexafluoro propylene)/polyethylene oxide based nanocomposite polymer electrolyte with ZnO nanofiller for dye-sensitized solar cell



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## ABSTRACT

Nanocomposite polymer electrolytes (NCPEs) were prepared by blending poly (vinylidene fluoride-co-hexafluoro propylene) copolymer (PVdF-HFP) and polyethylene oxide (PEO) polymers and incorporation of ZnO inorganic nanofiller (PVdF-HFP:PEO:EC:PC:NaI:I<sub>2</sub>:ZnO). The highest ionic conductivity value of 8.36 mS cm<sup>-1</sup> was recorded when 3 wt.% of ZnO inorganic nanofiller was incorporated into the NCPE system. Temperature-dependant ionic conductivity behaviour of NCPEs was analysed and proven to follow the Arrhenius thermal activated model. Structural studies of NCPEs were carried out using X-ray diffraction (XRD) and Fourier transform infrared (FTIR) spectroscopy analysis. NCPEs were used to fabricate Dye-sensitized solar cells (DSSCs). Enhancements in the solar light to electricity conversion efficiency ( $\eta$ ) of DSSCs were observed in the presence of ZnO inorganic nanofiller in the NCPE system and NCPE with 3 wt.% ZnO represented the highest  $\eta$  of 7.33% under full sun irradiation.

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## 1. Introduction

Dye-sensitized solar cells (DSSCs) are a new and low-cost technology which attracted the attention of many scientists. Electrolytes have a significant role in enhancing the performance of DSSCs [1–3]. Poly (vinylidene fluoride-co-hexafluoro propylene) (PVdF-HFP) and polyethylene oxide (PEO) blend polymers were used widely in the preparation of polymer electrolytes for DSSC applications [4,5]. GPEs are very safe for fabrication of practicable DSSCs and moreover, high ionic conductivity and well contact of GPEs with photo-anode and counter electrode reported to generate high-efficiency DSSCs [6]. Unique characteristics of PVdF-HFP and PEO polymers in GPE preparation, make them very suitable polymers for DSSC application. The strong interaction between CF<sub>2</sub> and C-O-C groups of PVdF-HFP and PEO respectively, helps in significant crystallinity reduction of GPE and high compatibility of these polymers for blending. PVdF-HFP provides a long-term stability of DSSC for practical usage due to its electrochemical stability beside

TiO<sub>2</sub> and Platinum (Pt) [7]. High electronegative Fluorine in PVdF-HFP and the high number of channels between electrodes of DSSC provided by PEO polymer helps in highly conductive GPE preparation as a result of fast ion transportations [8]. Low mechanical stability of GPEs affected by high plasticization of plasticizer in the system can be improved by using nanoparticles in system [9]. Nanoparticles increase amorphous phase and enhance ionic conductivity of GPEs by providing three-dimensional networks that separate polymer chains in the system hence enhances the mobility of I<sup>-</sup>/I<sub>3</sub><sup>-</sup> anions while passing through these free volumes obtained. Nanoparticles also decrease the recombination rate of electrons between the electrode and electrolyte interface in DSSCs [10–12]. ZnO has been used in many electrochemical devices as a wide-bandgap (WBG) semiconductor that possesses optoelectrical characteristic [13,14]. In this work ZnO, inorganic nanoparticles dispersed into the electrolyte system to produce nanocomposite polymer electrolytes (NCPEs) with enhanced ionic conductivity and mechanical properties. Electrochemical impedance spectroscopy (EIS) were applied to investigate the ionic conductivity and temperature-dependant ionic conductivity studies of NCPEs. Fourier-transform infrared (FTIR) and X-ray diffraction (XRD) spectra were obtained to investigate the interaction between materials and their molecular structure in NCPEs. DSSCs were

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fabricated by using NCPEs and their light-to-electricity conversion efficiencies were characterized under sun simulator.

## 2. Experimental

### 2.1. Materials

Poly (vinylidene fluoride-co-hexafluoro propylene) copolymer (PVdF-HFP), polyethylene oxide (PEO), ethylene carbonate (EC) and propylene carbonate (PC) were used in the preparation of NCPEs without any purification and as purchased from the Sigma-Aldrich. Zinc oxide nanopowder (<100 nm) was purchased from Aldrich. TiO<sub>2</sub> P90 (14 nm) and P25 (21 nm) were purchased from AEROXIDE and were used in preparation of electrode without any purification.

### 2.2. Nanocomposite polymer electrolyte preparation

Nanocomposite polymer electrolytes were prepared by mixing chemicals under continuously stirring and heating processes. Composition and relevant designation of the materials used in the preparation of nanocomposite polymer electrolyte are represented in Table 1. A clear mixture of ethylene carbonate (EC) and propylene carbonate (PC) as solvent and plasticizer with the same amount (4.0:4.0 g) was obtained by stirring under 60 °C for 1 h. For all the NCPEs, an optimized amount of sodium iodide (NaI) salt (0.70 g) was added to the mixture and dissolved by stirring for 4 h at the same temperature (60 °C). ZnO nanoparticles were dried in the oven for 4 h and under 60 °C before use. Certain amounts of ZnO nanoparticle were dispersed into each sample by heating and stirring for about 8 h. PVdF-HFP were separately dissolved in 10 ml of acetone by stirring at room temperature and the resultant solution added to the mixture gradually. Intended amount of PEO added to the mixture and the temperature adjusted to 80 °C and this mixture further stirred at this temperature for 4 h until evaporation of acetone in the mixture was obtained. At the end 0.118 g iodine (10 M of salt) was added to the mixture as a source of I<sup>-</sup>/I<sub>3</sub><sup>-</sup> redox couple and stirred until the homogeneous mixture was obtained. Electrochemical impedance spectroscopy (EIS) studies investigated directly after all NCPEs obtained and cooled down to the room temperature.

### 2.3. Dye-sensitized solar cell fabrication

Photo-anode preparation (anode electrode) technique followed by coating two layers of TiO<sub>2</sub> on the FTO-coated glass plates and according to our previous work [15]. The first and second layers of TiO<sub>2</sub> coated by two different methods of spin coating and doctor blade method respectively and after the photo-anodes obtained they immersed in the ruthenium-based N-719 dye solution for 24 h and dried. For counter electrode preparation, FTO-coated glass plates were coated with commercial platinum solution and sintered at 450 °C for 30 min. Dye sensitized solar cells fabricated by sandwiching each NCPE between photo-anode and counter electrode and their photovoltaic performance were investigated further.

### 2.4. Characterization method

Ionic conductivity and temperature-dependant ionic conductivity studied of NCPEs were carried out by using Hioki, 3532-50 LCR HiTESTER machine and under the frequency range of 50 Hz to 5 MHz the FTIR spectra for all NCPEs were obtained using ATR-FTIR PerkinElmer spectrum 400 spectrometer with minimum 0.4 cm<sup>-1</sup> spectral resolution. The XRD patterns were produced by PANalytical Empyrean diffractometer using Cu-K $\alpha$  radiation wavelength of  $\lambda = 1.540600 \text{ \AA}$  and in 2 $\theta$  range between 5° to 80° (45 kV and 40 mA). J-V curves for dye-sensitized solar cells fabricated by NCPEs were obtained under simulated sunlight ( $P_{in} = 1000 \text{ W m}^{-2}$ ) with multi-channel Autolab (PGSTAT30) potentiostat workstation.

## 3. Results and discussion

### 3.1. Ionic conductivity studies (EIS)

The equation represented below were used to calculate the ionic conductivity of NCPEs:

$$\sigma = \frac{L}{R_b A} \quad (1)$$

where  $\sigma$  is the ionic conductivity (S cm<sup>-1</sup>) of NCPEs, L is the thickness of the electrochemical cell filled with NCPEs,  $R_b$  is the bulk resistance and A is the area of the stainless steel blocking electrodes of the electrochemical cell. Fig. 1 represents the Cole-Cole plots for all the nanocomposite polymer electrolytes with different amounts of ZnO nanoparticles. Fig. 2 shows the variation of ionic conductivity values of NCPEs with ZnO nanoparticles content. Incorporation of ZnO nanoparticles into the NCPE system significantly enhanced the ionic conductivity up to 8.36 mS cm<sup>-2</sup> for PP-NaI-ZnO-3 in compare to the polymer electrolyte without ZnO nanoparticle (6.38 mS cm<sup>-2</sup>). It is inferred that ZnO nanoparticles provide three-dimensional channels for more ion transportations via Grotthuss mechanism, by increasing the amorphous phase of the NCPE system [16]. The ionic conductivity values for each NCPE are listed in Table 1. In addition to the improvements in ionic conductivity and the mechanical flow of the NCPEs, the contribution of ZnO nanoparticles enhances the electron recombination in the electrolyte by inducing electrons from conduction band of the TiO<sub>2</sub> layer on photo-anode to the oxidation/reduction potential of iodide/triiodide [17].

### 3.2. Temperature-dependant ionic conductivity studies

The ionic conductivity of NCPEs at different temperatures was obtained and the temperature-dependant ionic conductivity of NCPEs was investigated. The ionic conductivity of NCPEs increased by increasing temperature. Fig. 3 represents the temperature-dependant ionic conductivity behaviour of all nanocomposite polymer electrolytes with the incorporation of different amounts of ZnO nanoparticles. According to the unity of regression values ( $R^2 \sim 1$ ), it is concluded that NCPEs follow Arrhenius thermal

**Table 1**

Designation, ionic conductivity and activation energy values for nanocomposite polymer electrolytes with different amounts of ZnO nanoparticle.

Designation	[PVdF-HFP:PEO]:ZnO Composition (wt.%)	$\sigma$ , (mS cm <sup>-1</sup> )	$E_a$ , (meV)	Regression value ( $R^2$ )
PP-NaI-ZnO-0	[40.0:60.0]:0	6.38	98	0.99
PP-NaI-ZnO-1	[39.6:59.4]:1	6.92	97	0.99
PP-NaI-ZnO-2	[39.2:58.8]:2	7.30	96	0.98
PP-NaI-ZnO-3	[38.8:58.2]:3	8.36	95	0.99
PP-NaI-ZnO-4	[38.4:57.6]:4	6.73	97	0.99

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