Preparation and characterizations of PMMA-PVDF based polymer composite electrolyte materials for dye sensitized solar cell

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ABSTRACT

Blend polymer composite gel electrolytes are prepared using the polyvinylidene fluoride (PVDF), polymethyl methacrylate (PMMA) with alumina (Al₂O₃) in variance of alkali metal iodide salt system. The alumina doped blend polymer electrolytes characterized by the XRD diffraction and FT-IR spectra. This is supportive to the conformation of the crystallinity behaviour and the composite formation. The high-resolution scanning electron microscopy (HR-SEM) have used to find the composite electrolyte membrane porous size (10 μm) and it has support to understand the morphological structure of the membrane. To analyze the ionic conductivity of the potassium iodide based composite polymer electrolyte by the impedance measurements, which is 4.62 × 10⁻³ S cm⁻¹ at room temperature. Finally, different alkali metal iodide based dye-sensitized solar cells (DSSCs) fabricated and monitored an energy conversion efficiency.

1. Introduction

The dye sensitized solar cells (DSSCs) or Gratzel cells are much more interesting because it’s easy to fabricate and high energy conversion efficiency and comparatively low cost to other solar cells [1–3]. The standard DSSCs consist of dyes used as a sensitizer [4], nanocrystalline semiconductor metal oxide [5] coated onto transparent conducting glass plate act as a Photoanode, I₂/I⁻ redox couple containing electrolyte, platinum coated onto a glass plate used as counter electrode [6]. The liquid electrolyte assembled with electrode materials (cathode and anode) have been high energy conversion efficiency, even though it’s having some demerits of that as leakage of the electrolyte, evaporation of solvents, and photo-degradation of attached dye molecules and major drawback of corrosion of the counter electrodes [7–9]. This is contributing to a decrease in efficiency and the long-term stability of solar cells. Therefore, many attempts have been made to replace the liquid electrolytes by ionic liquids [10], organic-inorganic hole transport materials [11], solid polymer electrolytes [12], gel polymer electrolytes [13], blend polymer gel electrolytes and blend polymer composite electrolyte [14]. The gel polymer electrolytes is that higher ionic conductivity, excellent flexibility, higher efficiency and better stability for the DSSCs [15]. Even though, the blend polymer gel electrolyte or plasticized polymer electrolytes has some viscous nature, because, the ethylene and propylene carbonate [16] was added in the electrolyte preparation process, it has some viscous nature that has reduced the diffusion rate of I₃⁻/I⁻ redox couple. The lower diffusion rate of the redox couple affects the short circuit current density as well as reduced the efficiency of the DSSCs [17]. The blend polymer gel electrolytes were suitable and considerable interest in the recent generation of rechargeable batteries, super capacitors, dye sensitized solar cells and electrochromic device due to their excellent mechanical and thermal stability, flexibility and processability [18–21]. Even though, it has some drawbacks such as poor interface, contact between the dyes loaded TiO₂ photo-anode and counter electrode materials, lower I₋/I₃⁻ reproduction or regeneration capacity. In order to improve the light/photon-to-electrical energy conversion efficiency and long-term stability of the dye sensitized solar cells. To minimize their poor interfacial contact and faster regeneration of I⁻ ions having various approaches [22]. The blend polymer composite gel electrolyte was used in dye sensitized solar cell good interfacial contact between both the electrolyte-photoanode and electrolyte-cathode, decrease the crystalline nature of electrolyte membrane, improved the ionic conductivity and also increase the efficiency of the dye sensitized solar cells [23,24]. Higher light/photon-to-electrical energy conversion efficiency based on the dye sensitized solar cell was sufficient to faster dye regeneration of redox mediator sources. The electrolyte containing alkali metal iodide salts and iodine redox mediator that was faster dye regeneration compared to other redox mediator of Bromine/Bromide [25], cyanide/
thiacyanide redox couples of the electrolytes used in the DSSCs [26]. It is suffering from a few drawbacks such as poor interface contact between the dye loaded TiO2 photo-anodes and counter electrode materials, lower 1/3-reproduction or regeneration capacity. In this work, various alkali metal iodides salts based blend polymer composite gel electrolyte prepared with the alumina filler. Which are two different polymers were used; one is Poly methyl Methacrylate (PMMA i.e. optical transparency) and another one is Poly vinylene fluoride (PVDF). It has excellent optical properties with 99.9% transmission of visible light and filtration[27] and as well as it is possible to put on the PMMA polymer electrolyte membrane to near (front side) and lift (back side) illumination, PMMA used in DSSCs [28]. However, the ionic conductivity of this type of plasticized polymer films was offset by their poor mechanical properties at high concentration of plasticizers such as Ethylene carbonate (EC) and Propylene carbonate (PC). In order to overcome the problem of the poor mechanical strength of PMMA, It can be blended with various conducting polymers such as Polyethylene oxide (PEO) [29], Polyvinyl acetate (PVA) [30], Poly acrylonitrile (PAN) [31] and Poly vinylene fluoride-Hexafluoro phosphate (PVDF-HFP) [32] are used. PVDF has high electronegative fluorine. Conducting polymer was thermally stable even in presence of dye molecules attached to nano crystalline TiO2, and the platinum counter electrode in dye sensitized solar cell[33]. The filler materials and/or inorganic metal oxide nanoparticles like silica (SiO2), titanium dioxide (TiO2), alumina (Al2O3), zirconium dioxide (ZrO2) and lithium salts has been used in preparation of composite polymeric electrolytes for DSSCs [34–38]. These filler materials, improved the efficiency, reduced the Crystallinity, good interfacial contact stability between of both the electrolytes-anode and electrolyte cathode and also enhanced the ionic conductivity of composite gel electrolyte membrane. Among all metal oxides in this work Al2O3 has been used as a filler because its high thermal stability and also providing a large surface area and high conductivity [39,40]. The usage of blend polymer composite gel electrolytes in DSSCs shows a promising prospect to fabricate better DSSCs. The present work, the effect of the compositions on the properties of PMMA and PVDF was investigated using the Lithium and Sodium iodide salts with and without alumina is a filler material for the DSSCs. Because, PMMA is a high light transparency and is more impact resistant than glass, high ionic conductivity, excellent environmental stability, high solvent retention ability and good gelatinizing properties. In addition the PVDF is also thermomechanical properties materials due to semicrystalline nature, provides thermal stability while the amorphous phase (PMMA) provides the flexibility, for these reasons we chose these materials to the DSSCs.

2. Experimental

2.1. Materials

Poly(methylmethacrylate) (PMMA) and propylene carbonate were purchased from Hi-Media, poly(vinylene fluoride) (PVDF), ethylene carbonate, alumina nanopowder,1-hexyl-3-methylimidazolium iodide,hexachloro platinic acid, N719 dye, isopropanol, TiO2 (anatase) and carbonate, alumina nanopowder,1-hexyl-3-methylimidazolium iodide salts were purchased from Sigma Aldrich. Lithium and Sodium iodide salts were purchased from Alfa asar. Potassium iodide, ethanol and acetone were obtained from Merck Chemical Companies.

2.2. Preparation method

Blend polymer composite gel electrolytes were prepared by solvent casting method. Initially, 0.04 gm of alumina nano powder was well dispersed in 50 ml of acetone for 30 min by ultrasonically. To this solution was added 0.2 gm of PVDF and 0.2 gm PMMA, 0.04 gm LiI,ethylene carbonate, propylene carbonate 3:1 ratio, 0.04 gm of 1-hexyl-3-methylimidazolium iodide, 0.004 gm iodide was added under continuous heating with stirring to 80 C at 6 h. Finally viscous gel was transferred into dry pessl and allow for evaporation of acetone at room temperature. After evaporation of acetone, uniform thickness of films was obtained. The films were further dried to 40 °C at 12 h in a vacuum oven to remove trace amounts of acetone. The same procedure was used to prepare the NaI and KI based blend polymer composite gel electrolytes for further characterization.

2.3. DSSC fabrication

To prepare the dye sensitized solar cells, prepared the blend polymer composite electrolytes were used as an electrolyte for DSSc assembling, the photoanode preparation of TiO2 (anatase) was to grind the mortar with 3:1 vol ratio of ethanol and polyethylene glycol and then paste on the FTO glass plate by doctor blade technique. TiO2 coated FTO glass plate annealed at 450 °C then immersed in 0.2 M ethanolic using N719 dye solution for 12 h. After 12 h dye loaded TiO2 photo-anode was washed in ethanol solution to remove excess of dyes from anode surface. Counter electrode was prepared in 2-propanol and coated on FTO glass plate by spin coating method. The DSSCs were prepared by sandwiching type the blend polymer composite electrolyte between the dyes sensitized TiO2 and platinum counter electrode. Finally the active area of DSSCs was 0.5 cm × 0.5 cm.

2.4. Optimization conditions

During the fabrication of the DSSC device, primarily TiO2 nanoparticle and very few amount of PEG (binder) grind well and then coated on the fluorine doped tin oxide glass plate (FTO). In this TiO2 coated FTO glass annealed the 450 °C. After 3 h TiO2 coated on the FTO glass plate was washed with ethanol and TiCl4 solution. Ethanol used as trace amounts of untreated PEG removes the Photo-anode of TiO2/FTO and TiCl4 solution used as to prevent crack formation on the TiO2 coated on the Photo-anode. The preparation of the TiO2 coated on the FTO glass plate immersed in the 0.2 M solution of the N719 dye solution 12 h. After 12 h dye loaded on the FTO/TiO2 glass washed with small of amount of ethanol solution for the purpose of the removal of uncoated dye molecules. The platinum counter electrode was prepared from hexachloroplatinic acid 0.2 mM of hexachloroplatinic acid was dissolved in 2-propanol and spun coated on the FTO glass plate 2000 rpm for 5 min. DSSC was fabricated sandwich type of the PMMA-PVDF-KI-I2-EC-PC-Al2O3 composite electrolyte between the N719 dye sensitized FTO/TiO2 photo-anode and spin coated platinum counter electrode. Similar way DSSC fabricated the NaI and LiI based on the device. The active area is 0.5 cm2.

2.5. Characterization of composite gel polymer electrolytes

The crystal structures of the composite electrolytes were studied using XPer-tPro X-ray diffractometer with Cu-Kα radiation. Fourier transform infrared (FT-IR) spectra recorded at KBr pellets using Nicolet 5700 spectrophotometer (Thermo Electron Co. USA) in the wave number range 400–4000 cm⁻¹ determined the presence of functional group of three different alkali metal iodide in PVDF-PMMA-Al2O3 blend polymer composite gel electrolyte. The surface morphology of blend polymer composite electrolyte membranes was examined by scanning electron microscopy (HR-SEM) (FEI Quanta 250 Microscope, Netherlands). The electrochemical impedance spectra investigated by the PGSTAT 30 (Eco Chemi, Netherlands) in the range of 50 Hz to 1 MHz at amplitude of 10 million volts at room temperature. This instrument equipped with FRA2 software. The DSSCs performance was measured by using the solar simulator (150 W simulator, PEC-L11, PECELL), under air mass 1.5 and 100 mW cm² of the light intensity. The active area of the DSSC devices was measured by using a black mask of 0.5 cm². The photo-electrochemical parameters, i.e., the fill factor (FF) and light-to-electricity conversion efficiency(ηi), were
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