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Good prospect of ionic liquid based-poly(vinyl alcohol) polymer electrolytes for supercapacitors with excellent electrical, electrochemical and thermal properties

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ARTICLE INFO

Article history:

Received 29 November 2012

Received in revised form

26 April 2013

Accepted 14 June 2013

Available online xxx

Keywords:

PVA

Ionic liquid

Polymer electrolyte

Supercapacitor

Specific capacitance

ABSTRACT

Poly(vinyl alcohol) (PVA)/ammonium acetate ($\text{CH}_3\text{COONH}_4$)/1-butyl-3-methylimidazolium chloride (BmImCl) based polymer electrolytes were prepared by solution casting method. The ionic conductivity increased with temperature as shown in temperature dependent-ionic conductivity study. The maximum ionic conductivity of $(7.31 \pm 0.01) \text{ mS cm}^{-1}$ was achieved at 120°C upon adulteration of 50 wt% of BmImCl. The samples obeyed Vogel–Tamman–Fulcher (VTF) relationship. The glass transition temperature (T_g) of the polymer matrix was reduced by doping it with salt and ionic liquid as shown in differential scanning calorimetry (DSC). Supercapacitor was thus assembled. Wider potential stability range has been observed with addition of ionic liquid. Inclusion of ionic liquid also improved the electrochemical behavior of EDLC. The capacitance of supercapacitor were determined by cyclic voltammetry, electrochemical impedance spectroscopy and galvanostatic charge–discharge tester. The cell also illustrated energy density of 2.39 Wh kg^{-1} and power density of 19.79 W kg^{-1} with Coulombic efficiency above 90%.

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1. Introduction

As the world paces towards green technology, the implementation of environmentally friendly materials in manufacturing is an alternative way to reduce environmental issues of plastic waste disposal throughout all phases of the product life cycle. Therefore, biodegradable polymers have received an upsurge of interest nowadays in replacing those environmentally unfriendly polymers. Among biodegradable synthetic polymer, poly(vinyl alcohol) (PVA) is a multitasking material with excellent tensile strength and high ability to

form transparent film [1]. PVA has merits of being a host polymer due to its availability, good optical properties, cost effectiveness, non-toxicity, high hydrophilicity and good mechanical strength as well as high temperature resistance [2,3]. PVA also possesses some inherent characteristics, viz. biocompatible, ease of preparation, high abrasion resistance and high flexibility with superior chemical and thermal stabilities [4,5]. Other reasons for choosing PVA as host polymer are greater extent of polar groups (hydroxyl group) and high chain flexibility which promotes the salt-solvation [6]. High dielectric constant, excellent charge storage capacity and

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<http://dx.doi.org/10.1016/j.ijhydene.2013.06.061>

dopant-dependent electrical properties make PVA as a promising candidate in the development of electrochemical devices, especially EDLC and fuel cell applications [7]. Ammonium acetate is selected as doping salt in this current project. Ammonium acetate ($\text{CH}_3\text{COONH}_4$) is chosen due to its plasticizing effect. Besides, the acetate anion (CH_3COO^-) is preferred in salt-solvation through hydrogen bonding.

The PVA- $\text{CH}_3\text{COONH}_4$ polymer system has been widely investigated by G. Hirankumar and his co-workers [6–8]. However, the ionic conductivity is still too low and it is not applicable in the electrochemical devices. Therefore, several ways have been employed to improve the ionic conductivity, such as polymer blending, addition of plasticizers, impregnation of ionic liquids and doping of fillers. Among all the methods, addition of ionic liquid is the most viable technique to enhance the ionic conductivity because of its strong plasticizing effect, environmental friendly nature and high ion content. Ionic liquid is generally defined as a type of molten salt composed solely of ions. Other features of ionic liquid are excellent thermal stability, negligible vapor pressure, non-volatile, low melting point, non-flammable, low viscosity, wider electrochemical potential window and high ionic conductivity [9]. The objective of this present work is to prepare an environmentally friendly polymer electrolyte with superior electrical, thermal and electrochemical properties. Another aim is to investigate the effect of ionic liquid onto the polymer electrolytes and supercapacitors. In this paper, we report the effect of ionic liquid on the PVA- $\text{CH}_3\text{COONH}_4$ polymer electrolytes in supercapacitor application. Supercapacitor (or known as electrical double layer capacitor, EDLC) is fabricated using the highest conducting polymer membrane in this present work. Supercapacitor is the preferred choice compared to secondary batteries owing to the longer cycle life ($>10^5$ cycles), higher power density, higher capacitive density and faster charge-discharge rate as well as higher ability to be charged and discharged continuously without degrading [10].

2. Experimental

2.1. Materials

Polymer electrolytes containing PVA, $\text{CH}_3\text{COONH}_4$ and 1-butyl-3-methylimidazolium chloride (BmImCl) were prepared in this current work. Synthetic biodegradable polymer, PVA (Sigma-Aldrich, USA, 99% hydrolyzed with molecular weight of $130000 \text{ g mol}^{-1}$), $\text{CH}_3\text{COONH}_4$ (Sigma, Japan) and BmImCl (Acros organic, USA) were used as host polymer, salt and ionic liquid, respectively. All the materials were used as received.

2.2. Preparation of ionic liquid based-poly(vinyl alcohol) polymer electrolytes

Ionic liquid based-poly(vinyl alcohol) polymer electrolytes were prepared by means of solution casting. Table 1 shows the weight proportion of PVA, $\text{CH}_3\text{COONH}_4$ and BmImCl added into the polymer electrolytes and the designations of the polymer complexes. PVA was initially dissolved in distilled water. Appropriate amount of $\text{CH}_3\text{COONH}_4$ was subsequently

Table 1 – Nomenclature and weight percent of the materials added in the ionic liquid based-PVA polymer electrolytes.

Designations	Weight percent of PVA- $\text{CH}_3\text{COONH}_4$ -BmImCl		
	PVA (wt%)	$\text{CH}_3\text{COONH}_4$ (wt%)	BmImCl (wt%)
CL 0	70	30	0
CL 2	56	24	20
CL 5	35	15	50
CL 6	28	12	60

mixed in PVA solution. The weight ratio of PVA: $\text{CH}_3\text{COONH}_4$ was kept at 70:30. Different weight ratio of BmImCl was thus added into the PVA- $\text{CH}_3\text{COONH}_4$ mixture to prepare ionic liquid-based polymer electrolyte. The resulting solution was stirred thoroughly and heated at 70°C for few hours until a homogenous colorless solution was obtained. The solution was eventually casted on glass Petri dish and dried in an oven at 60°C . A free-standing polymer electrolyte film was obtained and shown in Fig. 1.

2.3. Characterization of ionic liquid based-PVA polymer electrolytes

2.3.1. Temperature dependence-ionic conductivity studies

Freshly prepared samples were subjected to ac-impedance spectroscopy for ionic conductivity measurements. Digital micrometer screw gauge was used to determine the thickness of the samples. The ionic conductivity of the polymer electrolytes was measured by HIOKI 3532-50 LCR HiTESTER impedance analyzer over a frequency range between 50 Hz and 5 MHz from ambient temperature to 120°C at a signal level of 10 mV. Polymer electrolytes were placed on the sample holder under spring pressure in the configuration of stainless steel (SS) blocking electrode/polymer electrolyte/SS electrode.

2.3.2. Differential scanning calorimetry (DSC)

DSC analysis was carried out by TA Instrument Universal Analyzer 200 which consists of DSC Standard Cell FC as main

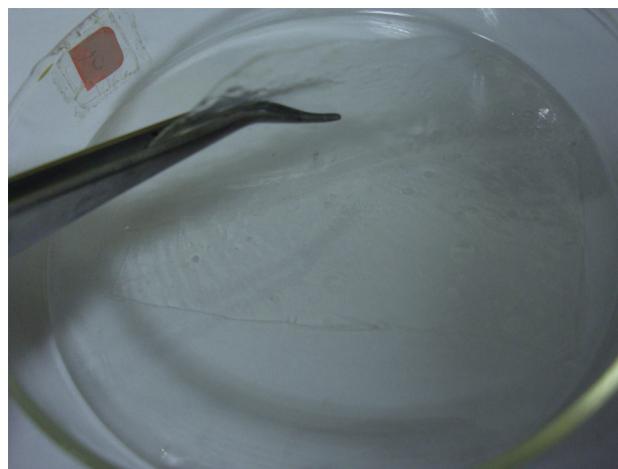


Fig. 1 – The image of ionic liquid-based PVA polymer electrolyte.

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