



Effect of alkyl-chain length of imidazolium based ionic liquid on ion conducting and interfacial properties of organic electrolytes



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ABSTRACT

In this study the effect of imidazolium ionic liquid with a different alkyl side chain (ethyl, propyl and butyl) on ion conducting and interfacial properties of organic electrolytes against carbon electrode has been investigated. The influence of electrochemical performances of imidazolium ionic liquids additives into organic solvent were examined by linear sweep voltammetry (LSV) cyclic voltammetry (CV), galvanostatic charge/discharge, and electrochemical impedance spectroscopy (EIS). The imidazolium ionic liquids with ethyl, propyl and butyl chains exhibited a variant electrochemical performance such as an improved specific capacitance, low resistance, good rate capability and excellent cycle ability. Among the ionic liquids, BMImBF₄ (10 vol.%)—containing electrolyte presented the maximum specific capacitance (130.5 F g⁻¹), capacitance retention (close to 80% after 1000 cycles) and low interfacial resistance of between electrolyte and electrode. It means that 10% BMImBF₄ in EC/DME (1/1) can be a proper electrolyte of capacitors.

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Introduction

Electrochemical double layer capacitors (EDLCs), also termed supercapacitors are promising energy storage device that the electrical charge in the electrochemical double layer was formed at the interface of electrode through the ion adsorption/desorption [1–5].

Activated carbons are the commercial electrode materials due to the high surface area, good conductivity and low cost [6]. The electrolytes used widely in a commercial capacitor are an aqueous solvent. Even though this device has many advantages such as high power and cycle ability, main disadvantage is low energy density ($E = 1/2 CV^2$) [7,8]. The energy density was related to the capacitance and operative voltage. To enhance the energy density, the capacitors should have wide potential window. Because the EDLC with aqueous electrolyte have low operative voltage, it is important to increase the potential window of device for a high energy density. However many investigations have been studied on the development of electrode materials, not for electrolyte materials [9,10,6].

Ionic liquids (ILs) which is called Room temperature molten salts composed of large organic cation and organic or inorganic anion. It stays in liquid state at room temperature. ILs have good

characteristics such as low melting points, chemical/thermal stability, non-flammability and large temperature range for liquid. Due to these features, they are considered as the most promising solvent for several applications of battery, fuel cells, and solar cells [11–15].

In this work, we investigated the usage of the imidazolium-based ionic liquids as additives in organic electrolyte (EC/DME) for EDLCs. The three imidazolium ILs with the different alkyl size have been selected for this work: 1-ethyl-3-methylimidazolium tetrafluoroborate (EMImBF₄), 1-propyl-3-methylimidazolium tetrafluoroborate (PMImBF₄) and 1-butyl-3-methylimidazolium tetrafluoroborate (BMImBF₄) as shown in Fig. 1. The effects of the molecular size and structure of electrolyte on the electrochemical double layer were analysed and the causes of these results were investigated when using the imidazolium ILs with different alkyl size as additives for capacitors.

Experiment details

Carbon electrode and electrolyte

The activated carbon (MSP-20, Kansai Coke and Chemicals, Japan) was used as active electrode materials. The carboxymethylcellulose (CMC)/styrene butadiene rubber (SBR) as a binder and carbon blacks as conductive agent were mixed with *N*-methylpyrrolidone (NMP) using the agate mortar for 30 min in glove box.

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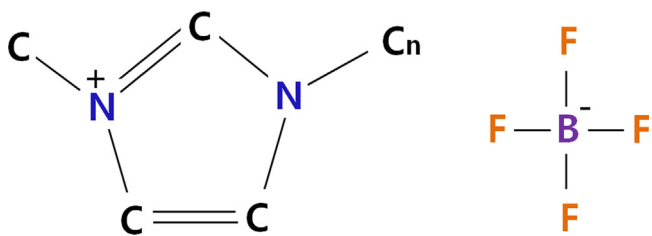


Fig. 1. Chemical structure of imidazolium cation with different alkyl size (C_n , $n = 2$; ethyl, $n = 3$; propyl, $n = 4$; butyl) and tetrafluoroborate anion.

The well-mixed slurry was coated on nickel foam ($1\text{ cm} \times 1\text{ cm}$) and dried at 100°C in the vacuum oven for 12 h. The electrode covered with slurry was then pressed. The average mass loading of active electrode materials was about 10 mg cm^{-2} .

The electrolyte was prepared by mixing EC (ethylene carbonate) and DME (dimethoxyethane) with 0.1 M TEABF_4 (tetraethyl ammonium tetrafluoroborate) in a glove box.

The imidazolium-type ionic liquids with different alkyl size such as 1-ethyl-3-methylimidazolium tetrafluoroborate (EMImBF₄), 1-propyl-3-methylimidazolium tetrafluoroborate (PMImBF₄) and 1-butyl-3-methylimidazolium tetrafluoroborate (BMImBF₄) were used as salts additives. The ionic liquids with the different volume ratio were also dissolved in prepared electrolyte to find the optimum content of additives

Measurement of the characterization of electrolyte

The ionic conductivities of electrolytes were measured by using the conductivity meter (ES-51, HORIBA, Ltd) at room temperature. The electrochemical stability of electrolyte was studied by linear sweep voltammetry (LSV) at scan rate of 20 mV s^{-1} in three electrodes system. The platinum wires were used as working/counter electrode and the Ag/AgCl reference electrode was used.

Evaluation of the electrochemical performance of EDLCs

The electrochemical behavior such as operative voltage, capacitance, cycle ability, galvanostatic charge/discharge and resistance was tested applying the cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) method.

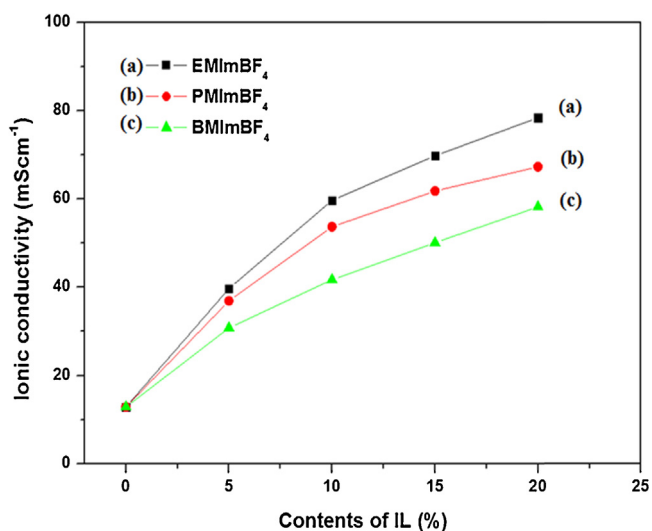


Fig. 2. Ionic conductivity changes as a function of imidazolium ionic liquids contents with ethyl, propyl and butyl in organic electrolyte.

The cyclic voltammetry (CV) measurements were carried out in three electrodes configuration that the prepared electrode was used as working electrode and platinum wire and Ag/AgCl electrode were used as counter and reference electrodes, respectively. The galvanostatic charge–discharge curves were plotted in operative voltage range at current density of 0.5 Ag^{-1} . The electrochemical impedance spectroscopy (EIS) method was determined by AC impedance spectroscopy over the frequency range from 100 kHz to 0.01 Hz with 5 mV amplitude modulation.

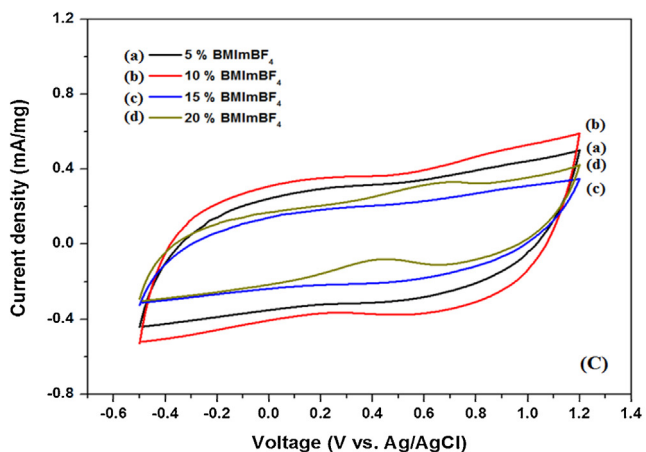
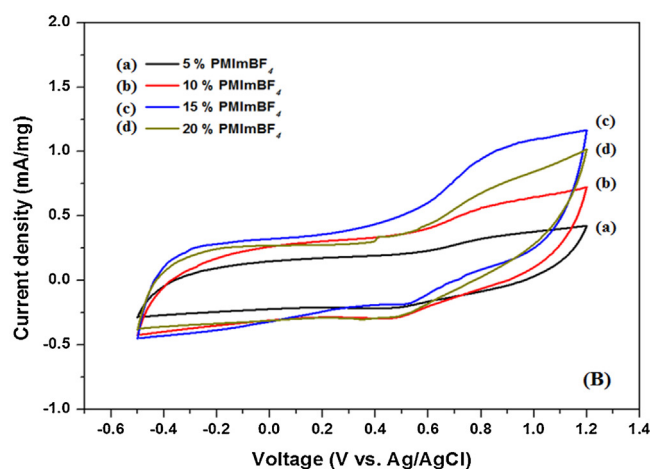
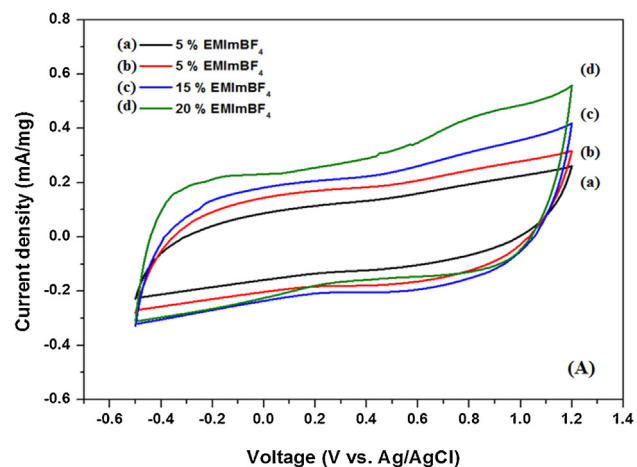


Fig. 3. Cyclic voltammograms at scan rate 5 mV s^{-1} of EC/DME electrolyte with 0.1 M TEABF_4 adding the different ratio of ionic liquids of (A) EMImBF₄, (B) PMImBF₄ and (C) BMImBF₄.

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