



Outstanding features of alginate-based gel electrolyte with ionic liquid for electric double layer capacitors



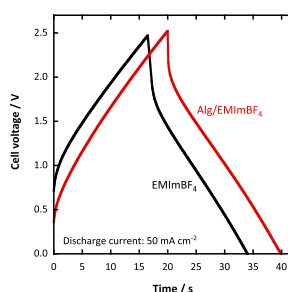
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HIGHLIGHTS

- An alginate (Alg)-based gel electrolyte is thermally stable.
- The Alg-based gel electrolyte is applicable for high voltage EDLC operation.
- Presence of Alg as a host polymer can significantly reduce the internal resistance of EDLC.
- High affinity of Alg to activated carbon delivers low resistive electrode/electrolyte interface.

GRAPHICAL ABSTRACT



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ABSTRACT

An alginate-based gel electrolyte with an ionic liquid (Alg/IL) is investigated for electric double-layer capacitors (EDLCs) by using physicochemical and electrochemical measurements. The Alg/EMImBF₄ (EMImBF₄ = 1-ethyl-3-methylimidazolium tetrafluoroborate) gel electrolyte is thermally stable up to 280 °C, where EMImBF₄ decomposes. Furthermore, the EDLC with the gel electrolyte can be operated even at high temperature. The cell containing Alg/EMImBF₄ is also electrochemically stable even under high voltage (~3.5 V) operation. Thus, the alginate is a suitable host polymer for the gel electrolyte for EDLCs. According to the result of charge–discharge characteristics, the voltage drop in the charge–discharge curve for the cell with Alg/EMImBF₄ gel electrolyte is considerably smaller than that with liquid-phase EMImBF₄ electrolyte. To clarify the effect of Alg in contact with the activated carbon electrode, we also prepared an Alg-containing ACFC electrode (Alg + ACFC), and evaluated its EDLC characteristics in liquid EMImBF₄. The results prove that the presence of Alg close to the active materials significantly reduces the internal resistance of the EDLC cell, which may be attributed to the high affinity of Alg to activated carbon.

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1. Introduction

Recent developments in energy storage devices that can deliver large amounts of energy at high power have addressed the

limitations in using mobile equipment, electric vehicles, and transportable systems, as well as in realizing small energy grids employing renewable energy sources. Electric double layer capacitors (EDLCs) are promising energy storage devices because of their high power density and long cycle lives, which are due to the presence of an electric double layer at the electrode–electrolyte interface [1,2]. Among the essential components of an EDLC cell, adequate electrolyte is necessary because it directly determines

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EDLC performances such as the rate characteristics and cycle life of the cell. Furthermore, to achieve high energy density, non-aqueous electrolytes are considered to be more favorable for EDLCs because of their wide electrochemical potential windows that provide high voltage operation, e.g., greater than 3 V [3].

Room-temperature ionic liquids (ILs) have attracted much attention as alternative electrolytes for EDLCs because of their unique properties, such as low volatility and low flammability, which can improve the safety of non-aqueous EDLCs [4–10]. The use of “liquid” IL electrolytes, however, may not be suitable for practical use when hazardous leakage is considered. Additionally, energy storage devices often require a solid-state electrolyte to construct not only high safety systems but also thin-film or stacked cells; hence, various gel-type electrolytes including ILs have been prepared [11–24]. Regarding EDLCs, various IL/polymer solid-state electrolytes have been proposed, thus far including poly (ethylene oxide) (PEO), poly (vinylalcohol) (PVA), poly (acrylonitrile) (PAN), and poly (vinylidene fluoride-co-hexafluoropropylene) (PVdF-HFP), which show relatively high ionic conductivity, and achieve long-term cycling stability [25–27]. However, few EDLCs with these gel electrolytes show specific capacitance and rate performance as high as those with liquid-phase ILs; this is most likely due to their lower ionic conductivity, thus hindering their practical application.

Recently, we proposed an alginate-based gel electrolyte with an IL (Alg/IL gel electrolyte) as a non-aqueous electrolyte for EDLCs [28]. Alg is a popular polysaccharide derived from brown seaweeds, is environmentally-friendly, non-toxic and highly abundant in nature, and has various practical uses (e.g., in food additives, as raw material for cosmetics, and in biochemical and medical products). Our previous study suggests that the proposed Alg/IL gel electrolyte is a promising electrolyte for EDLCs because its ionic conductivity is higher than those of other IL-based gel electrolytes. Furthermore, we observed that the EDLC with the Alg/IL gel electrolyte showed excellent cycle performance and high rate capability, comparable with the cell composed of a liquid-phase IL electrolyte. Such high performance may have originated from the effects of the high affinity of Alg toward the electrode materials [29], which results in an improvement at the electrode–gel electrolyte interface. We represent on here the extended follow-up study of the Alg/IL gel electrolyte to evaluate its thermostability and electrochemical stability because there is still concern over the thermal and the electrochemical deterioration of using a biopolymer. In addition, it is important to clarify the advantages of the presence of Alg in the gel electrolyte on EDLC performances. Our particular interest is the high rate capability of the EDLC cell with Alg/IL, probably attributed to the suitable interface between activated carbon electrodes and the gel electrolyte as described above. Thus, in this paper, an activated carbon fiber cloth electrode, whose pores are filled with the Alg/IL gel, is also prepared and evaluated to understand the advantages of using Alg giving an improvement of EDLC performance when Alg is more adjacent to the surface of the activated carbon fiber.

2. Experimental

2.1. Syntheses

The Alg/EMImBF₄ (EMImBF₄ = 1-ethyl-3-methylimidazolium tetrafluoroborate) gel film was prepared according to the same procedure reported previously [28]. Note that the content of the polymer matrix (i.e., Alg) in Alg/EMImBF₄ is approximately 10 wt.%, which is relatively low when compared with other conventional polymer electrolytes including 10–60 wt.% ILs [18,25,30–35]. To clarify how the presence of Alg is involved in the charge transfer reaction at the electrode–electrolyte interface, Alg-containing

activated carbon fiber cloth electrode (Alg + ACFC) was also prepared as described below. An Alg aqueous solution (3 wt.%), which was obtained by mixing sodium alginate (KIMICA ALGIN, KIMICA Corporation) and distilled water, was cast into an ACFC (ACC-562-25, Nippon Kynol Inc.; specific surface area: ca. 1950 m² g⁻¹). Then, the ACFC with sodium alginate was immersed in 1.0 mol dm⁻³ sulfuric acid as a crosslinking agent for a few minutes at ambient temperature. After gelation, the obtained composite was thoroughly rinsed with distilled water, and immersed in ethanol overnight. Next, the composite ACFC was punched out to prepare a disk that was 10 mm in diameter. The disk was rinsed again with ethanol, followed by immersion in EMImBF₄ (Kanto Chemicals Co., Ltd.). The Alg + ACFC was finally obtained by drying under vacuum for 48 h at 10⁻² Pa and 70 °C.

2.2. Measurements

Thermal analysis (TGA/DTG) of the Alg/EMImBF₄ gel electrolyte was evaluated using a thermogravimetric analyzer (Shimadzu, TGA-50). The measurement was performed under N₂ flow (50 mL min⁻¹) with a heating rate of 2 °C min⁻¹ and a temperature range from 50 to 400 °C. Ionic conductivity of the obtained Alg/EMImBF₄ gel electrolyte was evaluated via the AC impedance method using a computerized electrochemical interface (SI-128787A, Solartron Analytical) connected to an impedance/gain-phase analyzer (SI-1260, Solartron Analytical) with applied AC amplitude of 10 mV and frequency range from 500 kHz to 10 mHz. The ionic conductivity of the gel was estimated from the measured resistance when the imaginary component at high frequency fell to zero for a test cell fabricated with a pair of platinum disk electrodes and a gel sample that was mounted in a Teflon[®] container with platinum current collectors. To evaluate the temperature dependence of the ionic conductivity, the test cell was located in a thermally controlled chamber (SU-241, SPEC Co.).

The assembly of a two-electrode symmetric test cell with ACFCs and the gel electrolyte (Fig. 1a) is described in a previous report [28]. After a pair of ACFC electrodes was immersed in EMImBF₄ for

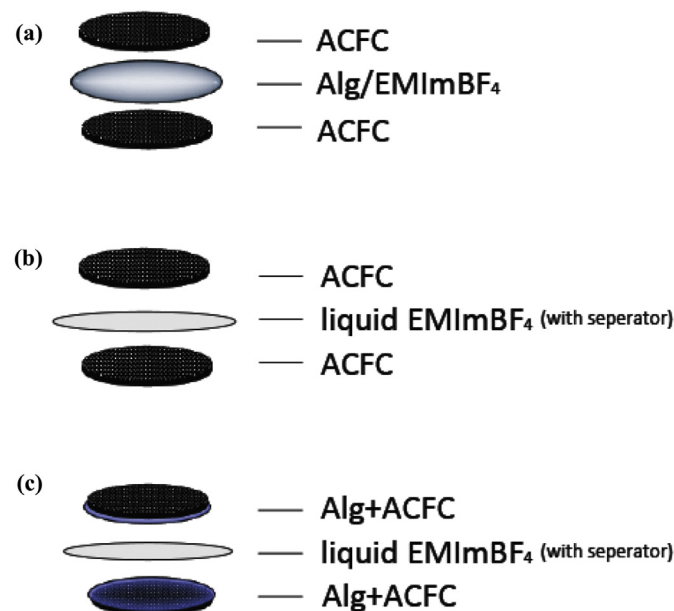


Fig. 1. Schematic illustrations of the EDLC test cells constructed in this study. (a) ACFC|Alg/EMImBF₄|ACFC, (b) ACFC|EMImBF₄|ACFC, and (c) Alg + ACFC|EMImBF₄|Alg + ACFC.

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