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Original Paper

Use of Natural Graphite for an Energy Storage Device

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Abstract

Ever growing high concerns over use of safe and low cost devices have provided a substantial attention on natural materials. As such natural graphite which has been deeply integrated into numerous applications is being received a consideration to be used for electrochemical devices. The main objective of this study is to explore the suitability of Sri Lankan natural graphite to serve in electrochemical double layer capacitors (EDLCs). In order to uplift the safety of the device, a gel polymer electrolyte was used instead of a liquid electrolyte. Two identical electrodes were consisted with Sri Lankan natural graphite as the active material and polyvinylidenefluoride as the binder. To prepare the electrolyte, polyvinylidenefluoride co hexafluoropropylene and magnesium perchlorate were used as the polymer and the salt respectively. Cyclic voltammetry test results show that single electrode specific capacitance is depending on the potential window. The percentage reduction of capacitance with continuous cycling was about 28%. Nyquist plot of EDLC further confirm the capacitive nature at low frequency.

Keywords

natural graphite, gel polymer electrolyte, electrochemical double layer capacitor, cyclic voltammetry, single electrode specific capacitance

1. Introduction

Graphite is one of the natural mineral resources in the world. It has been deeply integrated with diverse range of applications such as industries, transport, defence, medicine and sport. Graphite possesses remarkable features. It has high thermal resistance and high electric conductivity. Due to that graphite behaves as a metal and also as a non metal. Recently, the suitability of natural graphite to be served in electrochemical devices has been deployed mainly due to the reason that graphite structure is capable for ion incorporation which is of utmost importance for electrochemical devices.

Daunting array of energy and power demands have given rise to new insight for developing energy storage devices. Simultaneously, the global concerns over clean and low cost have given priority for those devices with eco friendly, naturally abundant materials. These issues have raised the motivation towards natural graphite (NR) very much.

Batteries and conventional capacitors had been the widely used energy storage devices for many years (Abruna et al., 2008). To meet the high rising energy demand, those two are not having a substantial capacity. As such, supercpacitors have been introduced which is renowned as a bridge to fill the gap between batteries and conventional capacitors (Kim et al., 2015). Supercapacitors are two fold namely electrochemical double layer capacitors (EDLCs) and redox capacitors. Former is based on carbon related electrodes whereas the latter uses conducting polymers and transition metal oxide electrodes.

Many groups have involved in fabricating batteries using graphite (Wang et al., 2017; Zhang et al., 2019; Lin et al., 2015). But, only a handful of research teams have employed graphite for EDLCs (Wang et al., 2012). From that portion, none is based on Sri Lankan natural graphite.

The main aim of the present study is to explore the performance of an EDLC fabricated using Sri Lankan natural graphite. As a measure of enhancing the safety, a gel polymer electrolyte has been used instead of a liquid electrolyte which has won a significant interest apart from the drawbacks.

2. Experimental

2.1 Preparation of NR Electrodes

NR sample received from Bogala Graphite Lanka (PVT) Ltd was used as received. It was first dissolved in isopropanol (Aldrich). 20% polyvinylidenefluoride (PVdF-Aldrich) was added into the mixture and sonicated well. Two fluorine doped tin oxide (FTO) glass plates of an area 1 cm^2 was cleaned and the slurry was coated on both. Then, they were allowed to dry at room temperature.

2.2 Preparation of the Gel Polymer Electrolyte (GPE)

Polyvinylidenefluoride co hexafluoropropylene (PVdF co HFP), magnesium perchlorate ($Mg(ClO_4)$) were used as received from Aldrich without any pre treatment. First, PVdF co HFP was dissolved in Acetone purchased from Aldrich. Mg (ClO_4) was mixed with the polymer solution and magnetic stirring was done for 24 hrs. The resultant was poured onto a petry dish and allowed solvent evaporation at room temperature.

2.3 Fabrication and Analysis of the EDLC

A GPE electrolyte sample having the identical shape and size of FTO glass plate electrodes was sandwiched in between the two electrodes. For the EDLC, cyclic voltammetry test was done varying the potential window using a computer controlled potentiostat (Metrohm Autolab M 101). Then, cycling was carried out using the same setup to observe the ability of EDLC to withstand for continuous charge discharge within the potential window, 0.1 V to 0.7 V. Impedance data were collected within the frequency range 0.01 Hz – 1 MHz using an impedance analyser (Metrohm Autolab M101).

3. Results and Discussion

3.1 Cyclic Voltammetry

Fig. 1 illustrates the cyclic voltammogrammes (CVs) obtained by varying the potential window.

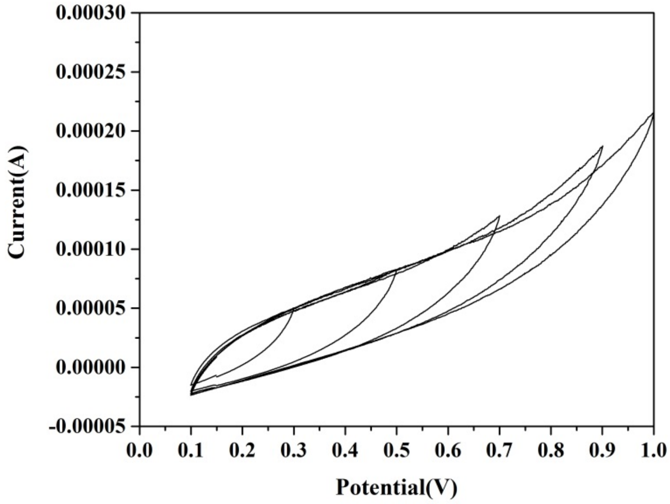

Figure 1. Cyclic Voltammogrammes Obtained with Widening Potential Window

When widening the potential window, the shape of CVs did not change very much. They were in near rectangle shape. This is an characteristic feature of EDLCs (Kim et al., 2015). But, at higher potentials, current increased tremendously. It may be due to any anodic reaction or electrolyte decomposition (Jayamaha et al., 2017). Single electrode specific capacitance, C_s was calculated using the following equation (Tey et al., 2016).

$$C_s = 2\int I dv / mS\Delta V$$

$\int I dv$ is the area of a CV, m is the single electrode mass, S is the scan rate and ΔV is the width of the potential window.

Continuous charge discharge performance of the EDLC is a good indication to exhibit the potential candidacy of an EDLC to be employed for practical applications. Figure 2 shows the variation of specific capacitance with the cycle number.

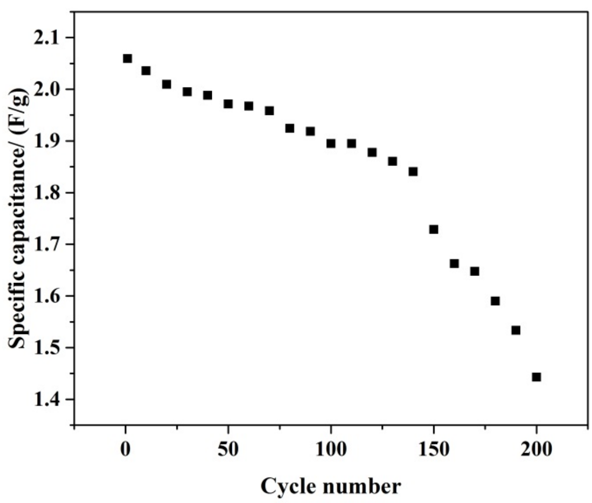

Figure 2. Specific Capacitance Variation with the Cycle Number

Charging discharging was done within the potential window from 0.1 V to 0.7 V. This might be possible with the use of a gel polymer electrolyte (Sun et al., 2012). Using an aqueous electrolyte, this is not possible. Anyway, it could be observed a reduction of C_s upon cycling. This has been reported by several groups (Yuan et al., 2006; Meller et al., 2014). The initial C_s of 2.05 F/g has reduced to 1.45 F/g during 200 cycles. The percentage of reduction is about 28% which is an encouraging value to carry forward further investigations to improve the performance.

3.2 Electrochemical Impedance Spectroscopy

Figure 3 is the resulted Nyquist plot obtained with the impedance data.

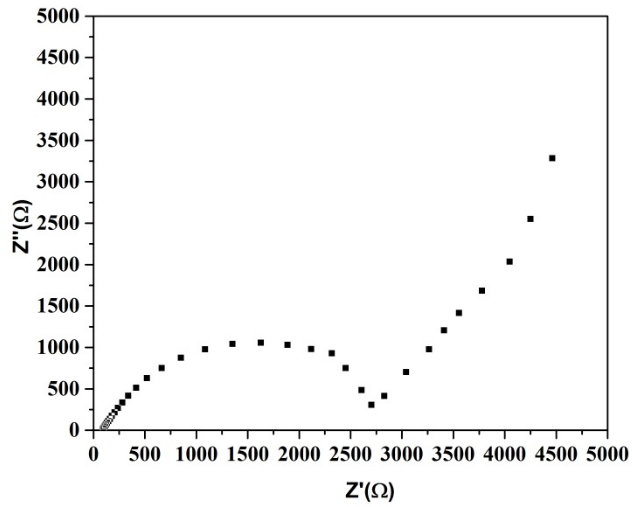

Figure 3. Nyquist Plot Obtained for the EDLC

Nyquist plots reveal many important properties of a device. Some of them are the bulk electrolyte resistance, charge transfer resistance and capacitive behavior (Fletcher et al., 2014). For an EDLC, the high frequency region of the Nyquist plots represents the resistive properties by semi circles. Spike in low frequency region shows capacitive properties (Prabaharan et al., 2006). For an ideal capacitor, the spike becomes parallel to the imaginary impedance axis. In the resulted figure, the semicircle that is relevant to the bulk electrolyte is absent due to the insufficient high frequency domain. The semicircle available in the plot is representing the charge transfer resistance. The tilted spike shows the capacitive properties (Prasadini et al., 2018). The tilted behavior may be due to some irregularities or roughness of the electrodes.

4. Conclusion

An EDLC was successfully fabricated using Sri Lankan natural graphite electrodes and a gel polymer electrolyte. This configuration is a novel attempt which is still in its infant stage. Cyclic voltammetry results as well as impedance data confer the suitability of the EDLC to be employed for applications after some developments.

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