

## Short Communication

# Graphene nanosheets as electrode materials for supercapacitors in alkaline and salt electrolytes



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## ABSTRACT

Carbon materials have played a significant role in the development of alternative clean and sustainable energy technologies. In particular, we will systematically discuss the applications of graphene nanosheets as an electrode material for supercapacitors. This article summarizes, the effect of size and nature of ions on pseudocapacitance and double layer capacitance of graphene electrode using CV and EIS techniques. The morphology and nature of the prepared electrode was investigated employing a scanning electron microscope. The prepared electrode shows better double layer characteristics in NaOH electrolyte in the potential range between  $-0.55$  and  $0.3$  V (V vs. SCE) at a scan rate of  $100$  mV s<sup>-1</sup>.

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## 1. Introduction

Electric double-layer capacitors (EDLCs), also called supercapacitors, are used in a wide range of energy capture and storage applications, and are believed to provide clean energy with nearly zero waste emission [1,2]. Compared with conventional capacitors, EDLCs can store much more energy for two main reasons: (1) a very small distance of charge separation at the interface between electrode and electrolyte and (2) larger amount of charges on the larger surface area of electrode [3]. The properties of the electrolyte system, solvent plus solute salt, required for supercapacitors

determine their electrical behavior [4]. Thus, the electrolyte plays an important role.

In this paper, the effects of ions size and specific properties on pseudocapacitance and double layer capacitance of the graphene electrode are investigated using CV and EIS techniques. The morphology and nature of the prepared electrode was investigated employing a scanning electron microscope.

## 2. Experimental

### 2.1. Materials

Graphene nanosheets (60 nm Flakes, multi-layered) with a specific surface area of  $15$  m<sup>2</sup>/g and purity of 98.5% were purchased from graphene supermarket and polytetrafluoroethylene ( $< 2$  μm) from Aldrich company. All other chemicals used in this study were purchased from Merck. In order to prepare the electrodes, a mixture containing 90 wt%

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graphene nanosheets (GNS) and 10 wt% polytetrafluoroethylene (PTFE) was prepared by mixing well in paste form in ethanol for about 30 min using ultrasonic wave. Paste form was chosen for homogenous dispersion of PTFE particles in the graphene matrix and to prevent the agglomeration of PTFE particles and graphene nanosheets. The main roles of a binder inside the electrode layer are: (1) to hold graphene nanosheets together forming a compacted porous layer; and (2) to help this electrode layer to adhere uniformly onto the current collector. Considering these properties, polytetrafluoroethylene (PTFE) has been recognized as one of the most popular choices. However, PTFE is a hydrophobic agent, and if too much PTFE is used, the porous electrode layer will become more hydrophobic, causing difficulty in both electrolyte penetration and ion mobility inside the matrix structure when an aqueous electrolyte is employed [5]. After drying the paste and powdering it, the composite was pressed onto a 316 L stainless steel plate (50 MPa) which served as a current collector (having a surface area of 1.4 cm<sup>2</sup>). A steel rod and a hollow cylinder of epoxy were used for pressing. The composite was pressed onto the epoxy properly by the steel rod. The typical mass load of the electrode material was 45 mg. The electrolytes investigated were 3 M NaOH and LiBr.

### 3. Characterization

Electrochemical characteristics of the electrode was determined by cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) in a three-electrode cell with 3 M NaOH and LiBr as electrolytes. The potential was cycled at different scan rates using a potentiostat (PGSTAT302N, Netherland) in the range of  $-0.55$  to  $0.3$  V. EIS measurements were also carried out in the frequency range of  $100$ – $0.02$  Hz at open circuit potential with an AC amplitude of  $10$  mV. Surface morphology and cross-section image of the electrode were examined with the scanning electron microscope (SEM, TESCAN, USA).

The specific capacitance can be estimated from the voltammetric charge surrounded by the CV curve according to the following formula [6,7]:

$$C = \frac{q_a + |q_c|}{2m\Delta V} \quad (1)$$

where  $q_a$  and  $q_c$  are the sums of anodic and cathodic voltammetric charges on positive and negative sweeps, respectively,  $m$  is the mass of active material (regardless of mass of PTFE) and  $\Delta V$  is the potential window of CV.

The real ( $C'$ ) and imaginary ( $C''$ ) capacitance of the electrode are calculated using the following equations [8,9]:

$$C' = \frac{Z''(\omega)}{\omega|Z(\omega)|^2} \quad (2)$$

$$C'' = \frac{Z'(\omega)}{\omega|Z(\omega)|^2} \quad (3)$$

where  $Z'(\omega)$  and  $Z''(\omega)$  are the respective real and imaginary parts of the complex impedance  $Z(\omega)$ ,  $\omega$  is the angular frequency and is given by  $\omega = 2\pi f$ .

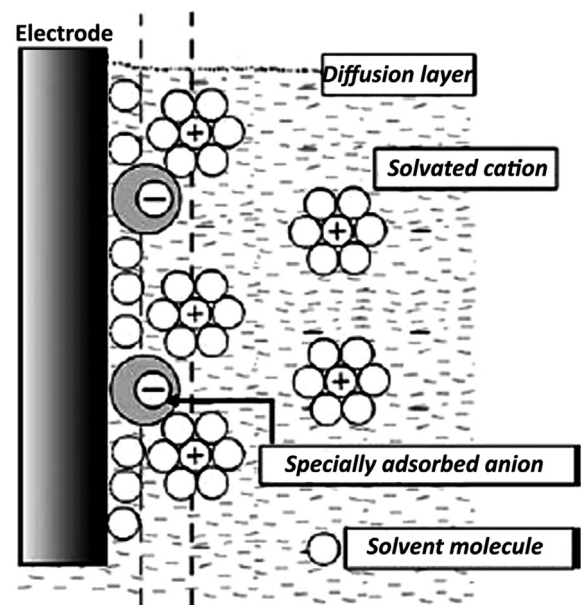


Fig. 1. Electrical double layer showing inner Helmholtz, outer Helmholtz and diffuse layer ( $\varphi_1$ =inner,  $\varphi_2$ =outer) [10].

### 4. Results and discussion

In stern model, the overall capacitance in EDLC ( $C_{dl}$ ), was considered as a series of capacitance,  $C_H$  and  $C_{diff}$ :

$$\frac{1}{C_{dl}} = \frac{1}{C_H} + \frac{1}{C_{diff}} \quad (4)$$

where  $C_H$  represents the compact Helmholtz layer formation from solvated ions attracted electrostatically in the OHP layer, while diffusion capacitance ( $C_{diff}$ ) results from the ions transportation, caused by a gradient between bulk and interfacial concentration of the electrolyte ions. Representation of the Helmholtz double layer is illustrated in Fig. 1. When the frequency increases or low conductive ions are used, the number of ions involved in the diffusion process can be reduced, therefore resulting in a decrease in capacitance. Graham's model presents the overall double layer capacitance which is composed of three components: adsorption capacitance ( $C_{ads}$ ), Helmholtz capacitance ( $C_H$ ) and diffusion capacitance ( $C_{diff}$ ) [11].

In Grahame's theory, it was recognized that dehydrated ions in IHP region could reside on the electrode surface with specific adsorption processes. This phenomenon results in adsorption capacitance,  $C_{ads}$ . In a certain system, this  $C_{ads}$  can be regarded as another capacitive element with some part of the electrochemical charge transfer process. The total capacitance ( $C_T$ ) can be represented by  $C_{ads}$  and in a series combination with  $C_{dl}$ :

$$\frac{1}{C_T} = \frac{1}{C_{ads}} + \frac{1}{C_{dl}} \quad (5)$$

It should be noted that two types of pseudocapacitance can arise in electrochemical processes: (1) adsorption pseudocapacitance and (2) redox pseudocapacitance [12]. However, it is necessary to differentiate  $C_{ads}$  from another pseudocapacitance which originates from Faradaic (oxidation/reduction

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