

# Carbide derived carbon electrode with natural graphite addition in magnesium electrolyte based cell for supercapacitor enhancements

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## Abstract

Herein, we have presented a supercapacitor based on carbide derived carbon (CDC) electrode with natural graphite (NG) addition. The capacitor was analyzed at 22 °C by cyclic voltammetry, galvanostatic charge-discharge and impedance techniques using a 0.5 mol/L of magnesium (II) bis (trifluoro methanesulfonyl) imide (MgTFSI) in ethylene carbonate-propylene carbonate (EC : PC = 1 : 1, v/v) as electrolyte. The results conclude that the CDC cell enhancements have been proven by the composite electrode (5%–30% NG to CDC) especially on the cell efficiency and voltage i.e., the CDC cell around 2.5 V limit was improved. An obtainable specific capacitance, real power and energy density are 15 F·g<sup>-1</sup>, 1.2 kW·kg<sup>-1</sup> and 15 Wh·kg<sup>-1</sup>, respectively.

## Key words

supercapacitor; carbide derived carbon; magnesium imide; organic electrolyte; natural graphite

## 1. Introduction

In recent years, an electric double layer capacitor (EDLC) also known as supercapacitor (SC) has been widely investigated for portable power and supporting device applications. The SCs are attractive due to their power density, capable of quick charge-discharge mode and long cell cycle life. Those properties are necessary for hybrid vehicles in which battery is a prime source [1]. This kind of the combined source is a green source of energy which is necessary for transportations. Also the SC has a vital role in modern electrical and electronics for immediate responses. The SCs are mainly driven by electrostatic [2] and faradic methods [3]. An organic medium plays a vital role in batteries [4] and SCs [5] due to their high potential limit and their excellent thermal stability [6]. Generally, the organic medium consists of propylene carbonate (PC), ethylene carbonate (EC) and dimethyl carbonate (DMC) solvents. The solvent system may be a single or mixture depends on their viscosity, dielectric constant, boiling and melting point requirements. For an example, conductivity of the EC/DMC mixture is higher than that of PC. But the PC is a better choice than the mixture for low temperature operation. The EC/PC mixture could be a potential candidate for a high operative potential window with good interfacial properties [7]. For a salt, imide salts have attractive electrochemical

characters such as non toxic, stable, conductivity with stable regions and stability at interfacial regions [8].

In order to minimize petrochemical utilities due to their air pollution [9,10], we need a power compensation for the petrochemical absentia [11,12]. In such cases, a high energy density SC might be necessary which can be reached by a high operative potential window. In terms of alternatives of Li and Na, also an equivalent nearly with the Li-Na systems, magnesium based system is the best choice. Unlike Li and Na based systems, the magnesium salts [2] are stable, safe, cheap, available (Mg is 13.9% abundant whereas lithium is ~10<sup>-3</sup>%), low toxicity and high negative potential with negligible hazard. The Mg systems are already proven as suitable electrolyte system in the field of energy storages also its electrochemical activity is comparable with Li based electrolytes. A low concentration leads to a better conductivity than that of Li salt. Many reports evidence the superior Mg electrolyte performances in aqueous [13], organic [1], ionic liquid [14] and gel forms [15].

On the SC electrodes view, porous carbon materials are proven as promising electrode materials due to their stability and wide range of electrochemical properties especially large surface area which can accommodate a huge solvated ions. Its properties depend on their surface area and its pore size distribution (PSD) [16]. So far, a numerous carbonaceous materials are recognized for the SC applications in which porosity is a

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crucial point of the carbon to the solvated ions because low dimension micropores reduce the ion insertions. Many carbon materials are analyzed on that above said point of view. Carbide derived carbon (CDC, Skeleton C, Oü Skeleton Technologies, Germany) is one of them. Some modified CDCs are also viewed for an efficient capacitors. Narrow PSDs, tunable PSD, high conductivity (0.16 S/cm) and stable voltage profile are some of the CDC important properties [17]. It also covers some practical advantages which include a high charge rate performance, a long cycle life due to lower functional group content on it and the pore accessibility for all media due to its narrow accessible mesopores [18–21], but the CDC meets an instability at high potential. Despite of the numerous investigations, the carbon applications are not limited, but growing in different directions like additive, composite and doping, for making an efficient capacitors. In order to improve a cell performance, an additive to a host carbon is a simple idea to minimize the drawback of the host carbon electrodes i.e. low conductivity, stability, electrolyte degrade. As we know, natural graphite is one of the most functionalized electrode materials for the lithium batteries [22]. Its high electronic conductivity might be useful for EDLC rather than its electrostatic character.

In this article, we have discussed a two electrode capacitor based on CDC, CDC-NG composite electrodes in the 0.5 mol/L of EC/PC/MgTFSI electrolyte. The role of NG in the CDC was studied by the cell performances. The cell status has also been studied by a symmetric and asymmetric configurations. Specific capacitance of the bare CDC cell was slightly decreased by the NG content, but a significant improvement was observed in charge rates and operative voltage limit. Finally, the contribution and the merit of NG in the CDC electrode were given.

## 2. Experimental

### 2.1. Materials

The solvents, propylene carbonate (PC) and ethylene carbonate (EC), from Sigma-Aldrich, were used without any further process. The 0.5 mol/L of magnesium (II) bis (trifluoro methanesulfonyl) imide salt (MgTFSI, Solvionic, France, 99.5%) in PC/EC (1/1 v/v) was taken as the cell electrolyte. The electrodes were prepared from a slurry of the carbide derived carbon (CDC) (SkeletonC, Oü Skeleton Technologies, Germany, surface area  $\sim 1450 \text{ m}^2 \cdot \text{g}^{-1}$ , effective surface area 882 ( $>0.8 \text{ nm}$ ), pore volume = 63 and micro pore volume  $\sim 28\%$ ) [23], carbon black (Alfa Aesar) as a conducting agent and polyvinylidene fluoride (( $\text{C}_2\text{H}_2\text{F}_2$ )<sub>x</sub>, PVDF, MW 540000 g/mol) as a binder at a weight ratio of 8 : 1 : 1 in *N*-methylpyrrolidinone ( $\text{C}_5\text{H}_9\text{NO}$ , NMP) as a dispersing agent. The slurry was coated on a stainless steel mesh of 90  $\mu\text{m}$  thickness with area of 1  $\text{cm}^2$ , and then it was dried overnight in vacuum at 120 °C. The active electrode material weight per cell was adjusted to  $\sim 3.5 \text{ mg}$  (carbon black and binder weights were excluded). A composite electrode was prepared in the above said way by adding natural graphite pow-

der (5  $\mu\text{m}$ , 12.1  $\text{m}^2 \cdot \text{g}^{-1}$ , SEC Co Ltd., Japan) to the CDC at 5, 10 and 30 wt%. A typical two electrode sandwich type cell was assembled using a cellulose separator of 120  $\mu\text{m}$  thickness in an argon glove box (M Braun,  $<1 \text{ ppm}$  of  $\text{O}_2$  and  $\text{H}_2\text{O}$ , room temperature of 25 °C). The prepared cells, electrode arrangements and indexes of the cells were as follows: (a) CDC|CDC cell (cell 1, C1), (b) NG5-CDC|NG5-CDC, i.e., 0.5% NG/7.5% CDC (cell 2, C2), (c) NG10-CDC|NG10-CDC (cell 3, C3), (d) NG30-CDC|NG30-CDC (cell 4, C4), (e) CDC|NG10-CDC (negative) (cell 5, C5) and (f) NG10-CDC (positive), CDC (cell 6, C6).

### 2.2. Techniques

The cell studies were performed with the help of a multichannel potentiostat workstation (EC-Lab V10.18, Biologic SA, VMP-3 France). The electrolyte stability was found from the linear sweep voltammetry (LSV) technique at a scan rate of 1  $\text{mV} \cdot \text{s}^{-1}$  in 0–6 V limit by the carbon electrodes. The electrochemical impedance spectra (EIS) of the cells were recorded at open circuit potential condition in a range of frequency from 100 kHz to 0.01 Hz with 2 mV amplitude. The corresponding specific capacitance  $C$  ( $\text{F} \cdot \text{g}^{-1}$ ) was calculated using the formula  $C = -1/(2\pi f Z'' m_t)$ , where  $Z''$  is the imaginary part of the complex impedance plot ( $\text{Im}$ , at 1 MHz frequency ( $f$ )),  $m_t$  is the weight of the two electrodes. The cell reversibility and symmetry were confirmed from the cyclic voltammetry (CV) experiments at  $\pm 2.5 \text{ V}$ . Also the CVs were checked in a range of 0 V to 3.0 V at scan rates of 2, 10 and 50  $\text{mV} \cdot \text{s}^{-1}$  for their electrostatic confirmation. Their gravimetric specific capacitance  $C$  ( $\text{F} \cdot \text{g}^{-1}$ ) was obtained from the formula  $C = i/(m_t v)$  where  $i$  is an average peak point of anodic-cathodic currents,  $m_t$  is the two electrode weights and  $v$  is the scan rate. Finally, the cell capacitance and cycling behaviors were tested by galvanostatic charge-discharge (CD) studies. The CD studies were carried out from 0 V to 3 V limit at various current densities of 100, 200, 500 and 1000  $\text{mA} \cdot \text{g}^{-1}$ . The corresponding specific capacitance  $C$  ( $\text{F} \cdot \text{g}^{-1}$ ) was calculated according to the relation  $C = I t / V m_t$ , where  $I$  is applied current,  $t$  is discharge time in s,  $m_t$  is total electrode weights and  $V$  is the potential window. Additionally, the real energy density and power density of the cell were calculated for the cell evaluation. The energy density relation is  $E_{\text{real}} = 1/2 C V^2$ , where  $C$  is the specific capacitance from the CD studies, and  $v$  is the potential limit ( $E_{\text{real}}$  means IR drop included). The real power density is obtained from the relation  $P_{\text{real}} = E_{\text{real}} / t_{\text{Discharge time}}$ , whereas a maximum power density can be obtained from the relation,  $P_{\text{max}} = V_i^2 / 4R$ , where  $V_i$  = potential window,  $R$  = equivalent series resistance (ESR).

## 3. Results and discussion

### 3.1. Linear sweep voltammetry (LSV)

The electrolyte stability was checked by LSV technique using a two electrode CDC cell. The LSV result gives an approximation on the electrolyte working potential limit. The

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