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Tsay, Keh-Chyun; Zhang, Lei; Zhang, Jiujun

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Effects of electrode layer composition/thickness and electrolyte concentration on both specific capacitance and energy density of supercapacitor

Keh-Chyun Tsay, Lei Zhang*, Jiu-Jun Zhang

Institute for Fuel Cell Innovation, National Research Council of Canada, Vancouver, BC V6T 1W5, Canada

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ABSTRACT

In this paper, the effects of several experimental conditions, such as electrode layer binder content, conducting carbon content, electrode layer thickness, as well as electrolyte concentration, on both the specific capacitance and energy density of a BP2000 carbon-based supercapacitor are investigated using both cyclic voltammetry and a galvanic charging–discharging curve. The electrode layer studied contains Super C45 carbon as the conducting additive, PTFE as the binder, and Na_2SO_4 as the aqueous electrolyte, respectively. With the purpose of optimizing the electrode layer structure, 15 wt% of Super C45 and 5 wt% of PTFE in the electrode layer with a thickness of 100 μm , are found to be the best composition in terms of improving both specific capacitance and energy density. Regarding the effect of electrolyte concentration in the range of 0.1–1.0 M, 0.5 M of Na_2SO_4 gives the best performance.

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1. Introduction

In recent years, electrochemical supercapacitors (ESs) have been recognized as key energy efficiency devices for rapid energy storage and delivery. This is due to their high power density, long lifecycle, high efficiency, wide range of operating temperature, environmental friendliness, and safety, as well as serving as a bridging function for the power/energy gap between traditional dielectric capacitors (which have high power output) and batteries/fuel cells (which have high energy storage) [1,2]. With these advantages, ES have become very competitive for applications such as electric hybrid vehicles, digital communication devices such as mobile phones, digital cameras, electrical tools, pulse laser technique, uninterruptible power supplies, and storage of the energy generated by solar cells [3]. However, several challenges such as low energy density, high cost, and high self-charge rate have limited its wider applications.

Regarding low energy density, one of the major limitations is induced by the low specific capacitance of the electrode materials used. At the current state of technology, the most practical material for ES electrode layer is nano-scaled carbon, such as activated carbon, carbon aerogels, carbon nanotubes, templated porous carbons and carbon nano fibers. Although some hybrid materials between carbon and metal oxides such as MnO_x and RuO_x have been used

to construct EC electrode layers, they are still in a stage of research and development [3]. Therefore, nano-scaled carbons are still the preferred ES electrode materials.

In general, the amount of energy stored in nano-scaled carbon-based supercapacitors is determined by the specific capacitance of the carbon, the electrical/ionic conductivity of the electrode layer, the ionic conductivity of the electrolyte filled separator, as well as the voltage window of the electrolyte [4]. Regarding specific capacitance of the electrode layer, specific surface area, pore size, and electrode structure of electrodes are dominating factors [5–8]. Two carbon surfaces exist in the electrode layer: one is the external surface and the other is the inner surface, and each surface can give their corresponding specific capacitances [9–12]. It follows both surfaces make contributions to the total specific capacitance. From our experiments with Carbon black BP2000-based material, we observed that the contribution from the external carbon surface to the total capacitance is around 40% and the other 60% contribution came from the inner microporous surface. Barbieri et al. [13] studied the relationship between specific capacitance of various active carbon materials and their specific surface areas, and found that the specific capacitance could increase linearly with specific surface area within a specific range of surface areas. Shi et al. [5] also found a linear relationship between specific capacitance and surface area. These results indicate that carbon surface area plays an important role in increasing specific capacitance.

In addition to using carbon material with high surface area, for practical purposes, type of binder and its concentration, thickness of the electrode layer, as well as preparation procedure are also

* Corresponding author. Tel.: +1 604 221 3000x5504; fax: +1 604 221 3001.
E-mail address: lei.zhang@nrc.gc.ca (L. Zhang).

important in achieving high specific capacitance, and in turn high supercapacitor performance. The purpose of this work is to optimize the supercapacitor's performance by investigating the effects of these factors on the electrode layers.

In this paper, we report our attempt in the optimization of carbon based supercapacitor performance using in-house fabricated electrode layers and modified test cell apparatus. Several factors affecting electrode preparation were investigated including binder content, conducting carbon content, as well as electrode layer thickness. Electrochemical methods such as cyclic voltammetry (CV) and galvanic charging-discharging curve (GCC) were employed for testing electrode performance.

2. Experimental

2.1. Materials for electrode layer

For electrode layer preparation, Carbon black BP2000 (Cabot Corporation), conducting carbon Super C45 and Super P Li (both from TIMCAL), and 60 wt% polytetrafluoroethylene (PTFE)-water dispersion (Aldrich) were used as received without further modification.

2.2. Electrode layer preparation

For electrode layer preparation, carbon and conducting carbon powders were mixed with a Vortex Mixer (Thermo Scientific) for 30 min to form a uniformly mixed power. This powder was then transferred into a beaker containing both PTFE binder and ethanol solution under constant stirring to form a powder suspension. Then the temperature of this suspension was raised to evaporate the solvent. After most of the ethanol was removed, the paste formed was collected on a glass plate. Using a spatula, this paste was manipulated by repeatedly folding and pressing till a sufficient mechanical strength was achieved. Then this paste was rolled to a thin electrode sheet with the required thickness using a MTI rolling press. Finally, this electrode sheet was placed into a vacuum oven at 90 °C under active vacuum for at least 12 h.

For electrode layer preparation, the dry electrode sheet was cut into two 2 cm × 2 cm electrode layer squares. Then these two electrode layers were sandwiched with a 30 μm thick porous PTFE separator (W.L. Gore & Associates, Inc.) in the middle, forming an electrode separator assembly (ESA). The assembly was then impregnated in an aqueous electrolyte containing Na₂SO₄ with desired concentration inside a vacuum oven at 60 °C for at least 1 h. This electrolyte-impregnated ESA was then assembled into the two-electrode test cell purchased by IMI and modified in our laboratory.

2.3. Two-electrode test cell assembling and electrochemical measurements

Fig. 1 shows the two-electrode cell design for testing and diagnosing supercapacitor performance. The active electrode surfaces for both positive and negative electrodes are all 4.0 cm². It can be seen that the two electrodes are placed between two Teflon plates with two metal plates outside. These two metal plates serve as holders to tighten the ESA together. On one metal plate, there are three screws (M5 × 0.8 mm) with a three-point geometry, which ensure a better pressure balance than a four-point geometry. Between each Teflon plate and electrode there is a metal sheet serve as current collector. During assembly, screws were tightened with a torque screw driver to 80 cN m. The pressure from the stainless steel plates can be directly transferred to the ESA through two stainless steel coins placed on the back of each Teflon plate of the internal cell. The test cell was then set in a 100 mL volume beaker filled with

about 20 mL of Na₂SO₄ solution. The beaker together with the test cell and electrolyte were placed in a vacuum oven at 60 °C for at least half an hour to remove trapped air.

For electrochemical measurements, cyclic voltammograms and galvanic charging-discharging curves were recorded using a Solartron 1287 potentiostat. For cell internal resistance measurements, a Tsuruga 3566 AC m-ohm Tester was used at an AC frequency of 1 kHz. All measurements were made at room temperature and ambient pressure.

2.4. Specific capacitance calculation based on the experiment measurements

Since the two electrodes are identical and the electrode material is Carbon black BP2000, which is non-faradic, the supercapacitors we studied in this paper should be symmetrical and pure double-layer supercapacitors. For a symmetrical double-layer supercapacitor, the capacitance measured from cyclic voltammogram (CV) or galvanic charging-discharging curve (GCC) in a full cell hardware should be calculated according to Eqs. (1)–(4) [14]:

$$\frac{1}{C_T} = \frac{1}{C_p} + \frac{1}{C_n} \quad (1)$$

where C_T is the total capacitance (F) measured using the two-electrode cell apparatus, C_p is the capacitance of the positive electrode layer, and C_n is the capacitance of the negative electrode layer. For a symmetric supercapacitor, C_n should equal to C_p . If we can define the individual electrode capacitance (C):

$$C = C_p = C_n \quad (2)$$

Eq. (1) will become Eq. (3):

$$C_T = \frac{1}{2}C \quad (3)$$

For a positive or negative electrode, the specific capacitance of the active material (C_S) of this electrode can be defined as:

$$C_S = \frac{C}{m_{act}} = \frac{2C_T}{m_{act}} \quad (4)$$

where m_{act} is the mass weight of active material in the positive or negative electrode layer. Note that this Eq. (4) is only applicable for a symmetric supercapacitor. From Eq. (4), it can be seen that the specific capacitance can be obtained by measuring the total cell capacitance (C_T).

The total capacitance C_T can be calculated using the measured charge quality Q , which is either the total charge at forward charging direction (Q_f) or that at the backward charging direction (Q_b) in the cell voltage range of V_1 to V_{cell} . If V_1 is the initial cell voltage, and summing it equals to 0, and V_{cell} is the voltage at a fully charged state. Normally, $Q_f = Q_b$. Therefore, C_T can be expressed as Eq. (5):

$$C_T = \frac{Q}{V_{cell}} = \frac{Q_f}{V_{cell}} = \frac{Q_b}{V_{cell}} \quad (5)$$

Note that from the CV curve, the charges Q_f and Q_b can be obtained through integration, as expressed by Eqs. (6) and (7):

$$Q_f = \int_{V_1}^{V_{cell}} I(V)dt = \int_{V_1}^{V_{cell}} I(V)v \, dV \quad (6)$$

$$Q_b = \int_{V_{cell}}^{V_1} I(V)dt = \int_{V_{cell}}^{V_1} I(V)v \, dV \quad (7)$$

where v is the voltage scan rate, $I(V)$ is the current, which is a function of cell voltage (V), dV and dt are the cell voltage and time changes with the voltage scanning, respectively. In a similar way,

Fig. 1. Schematic illustration of the two-electrode supercapacitor test cell.

from the galvanic charging–discharging curve (both the charging current, I_f , and discharging current, I_b , are constant), the charges Q_f and Q_b can also be obtained through integration, as expressed by Eqs. (8) and (9):

$$Q_f = \int_0^{t_{\text{end}}} I \, dt = I_f t_{\text{end}} \quad (8)$$

$$Q_b = \int_0^{t_{\text{end}}} I \, dt = I_b t_{\text{end}} \quad (9)$$

where t_{end} is the charging or discharging time when the supercapacitor is fully charged or discharged.

2.5. Energy density calculation based on the measured cell voltage and specific capacitance

In order to obtain the expression of energy for a supercapacitor, Fig. 2 schematically shows the charging curve of a double-layer supercapacitor at a constant charging current of I_f . Assuming that

Fig. 2. Schematic illustration of the charging curve of a pure double-layer supercapacitor (the equivalent series resistance (ESR) and parallel leakage resistance are not considered here).

the cell is fully charged at a time of t_{end} , giving a cell voltage of V_{cell} , as shown in Fig. 2, the energy (E_{SC}) can be expressed as:

$$E_{\text{SC}} = Q_f V = \int_0^{t_{\text{end}}} I_f \, dt V = I_f \int_0^{t_{\text{end}}} V \, dt \quad (10)$$

From Fig. 2, it can be seen that the area under the charging line is marked by S_1 , and the area above the charging line is marked by S_2 .

The term of $\int_0^{t_{\text{end}}} V \, dt$ in Eq. (10) is equal to S_1 , which can be expressed as Eq. (11):

$$\int_0^{t_{\text{end}}} V \, dt = \frac{S_1}{S_1 + S_2} V_{\text{cell}} t_{\text{end}} \quad (11)$$

Combining Eq. (11) with Eq. (10), we can obtain:

$$E_{\text{SC}} = \frac{S_1}{S_1 + S_2} I_f t_{\text{end}} V_{\text{cell}} = \frac{S_1}{S_1 + S_2} Q_f V_{\text{cell}} \quad (12)$$

Combining Eq. (5) with Eq. (12), a general expression for energy can be obtained:

$$E_{\text{SC}} = \frac{S_1}{S_1 + S_2} C_T V_{\text{cell}}^2 \quad (13)$$

For a pure double-layer supercapacitor, the charging curve is a straight line, so $S_1 = S_2$, suggesting that $S_1/(S_1 + S_2) = 1/2$. Therefore, Eq. (13) can be written as Eq. (14):

$$E_{\text{SC}} = \frac{1}{2} C_T V_{\text{cell}}^2 \quad (14)$$

This equation has been widely discussed and used in literature [15]. It should be noted that this Eq. (14) is only applicable to the case of ideal double-layer supercapacitors without equivalent series resistance and parallel leakage resistance. If the electrode material is Faradaic or hybridized between double-layer and Faradaic materials, the charging curve may not be approximated to a straight line, so that S_1 is not equal to S_2 . In this case, the factor $S_1/(S_1 + S_2)$ will not equal to 1/2, Eq. (14) will not be accurate anymore.

Fig. 3. (a) Cyclic voltammograms, recorded at various voltage scan rates using a carbon BP2000-based supercapacitor with an electrode composition of BP2000:Super C45:PTFE = 80:15:5 (wt%), electrode thickness of 100 μm , and active carbon loading of 3.0 mg/cm^2 . (b) Specific capacitance as a function of voltage scan rate.

Furthermore, the energy density of the active electrode material ($E_{\text{SC-act}}$) can be obtained by dividing C_T in Eq. (13) by the mass weight of active electrode material m_{act} :

$$E_{\text{SC-act}} = \frac{1}{2} \frac{C_T}{m_{\text{act}}} V_{\text{cell}}^2 \quad (15)$$

If the individual electrode capacitance (C) is obtained by CV method, the energy density of the active electrode material can be expressed alternatively based on Eqs. (3) and (15):

$$E_{\text{SC-act}} = \frac{1}{4} \frac{C}{m_{\text{act}}} V_{\text{cell}}^2 = \frac{1}{4} C_S V_{\text{cell}}^2 \quad (16)$$

Again, this equation is only applicable to the case of pure double-layer supercapacitors. For a Faradaic or hybrid supercapacitor, Eq. (13) should be used.

3. Results and discussion

3.1. Specific capacitance measured by cyclic voltammetry and the effect of potential scan rate on specific capacitance

Fig. 3(a) shows the cyclic voltammograms of a symmetrical supercapacitor cell whose both electrodes were composed of BP2000 carbon particles. The electrode layer consisted of BP2000 carbon, Super C45 and PTFE with a wt% ratio of 80:15:5. It can be seen that at low scan rates, the CVs display an ideal capacitive behaviour (rectangular shape), but when the scan rate is increased, this ideal behaviour is distorted with a gradual loss in cell specific capacitance which is measured and calculated according to Eqs. (1)–(4). For more clarity, Fig. 3(b) shows the dependency of specific capacitance on potential scan rate. The observed decrease in

Fig. 4. (a) Charging–discharging curves, recorded at various charging rates using a carbon BP2000-based supercapacitor with an electrode composition of BP2000:Super C45:PTFE = 80:15:5 (wt%), electrode thickness of 100 μm , and active carbon loading of 3.0 mg/cm^2 ; (b) specific capacitance as a function of charging rate; and (c) energy density as a function of charging rate.

specific capacitance with increasing the potential scan rate is due to the limited transfer of ions to the carbon particle surface, resulting in inaccessible pore portions of the electrode layer at high scan rates (charging rates). This phenomenon is typical for all types of supercapacitors, reflecting the limited mass transfer kinetics within the porous electrode layer [16–18].

3.2. Specific capacitance and energy density measured by cell charging–discharging curves, and the effect of charging rate on the performance

Fig. 4(a) shows the charging–discharging curves at the charging rates from 5 mA to 100 mA in the cell voltage range between 0 and 1.0 V. This charging–discharging current range covers nearly the same range in cyclic voltammogram measurements except 2 and

Fig. 5. Specific capacitance and energy density of carbon BP2000 electrode material as a function of PTFE contents. Data from Table 1.

5 mV/s which have charging/discharging currents less than 1 mA. The specific capacitances at various charging–discharging currents were calculated based on the measured C_T , V_{cell} , and Q using Eq. (5), and plotted as a function of charging–discharging current, as shown in Fig. 4(b). Based on these specific capacitance values, the energy density can be calculated according to Eq. (16), and plotted as a function of charging–discharging current, as in Fig. 4(c). Specific capacitance obtained from low charging rates are slightly higher than those from high rates. This is due to limited ion accessible time to the micro pore [6]. The average particle size reported is about 12–15 nm [19,20] and some micro pores with size less than 4 nm contribute a significant portion of pore volume according to our BET measurements. Smaller micro pores are more difficult to access and require the charging–discharging rate to be lower than a critical value. When the charging–discharging rate is higher than this critical value, current perturbation is unable to reach inside the pore.

3.3. Effect of binder content

The main roles of a binder inside the electrode layer are: (1) to hold carbon particles together forming a compacted porous layer; and (2) to help this electrode layer to adhere uniformly onto the current collector. Materials with a three-dimensional network structure linked by long molecular chains, and good electrochemical stability as well as good adhesion to metal plate are desired for electrode layer binders. With respect to this, 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,

Fig. 6. Specific capacitance and energy density of carbon BP2000 electrode material as a function of conducting carbon content. Data from Table 2.

Table 1
Performance data of Carbon black BP2000-based supercapacitor, measured by both cyclic voltammetry (at a potential scan rate of 20 mV/s), and galvanic charge–discharge curves (at a charging–discharging rate of 5 mA/cm²). The electrolyte: 0.5 M Na₂SO₄ aqueous solution (Room temperature); conducting carbon (Super C45) content in the electrode layer: 15 wt%; and electrode layer thickness: 100 μm.

Binder content (wt%)	Active carbon loading, m_{act} mg/cm ²	Specific capacitance from CV, C_s (F/g active carbon)	Electrode geometric capacitance from CV, C' (F/cm ²)	Specific capacitance from GC, C_s (F/g active carbon)	Electrode geometric capacitance from GC, C' (F/cm ²)	Average specific capacitance, C_s (F/g active carbon)	Cell voltage, V_{cell} (V)	Energy density, $E_{spc-act}$ (Wh/kg active carbon)
3.0	2.95	80.0	0.118	82.3	0.121	81.2	1.00	5.64
5.0	3.00	86.1	0.129	88.2	0.132	87.2	1.00	6.05
8.0	2.73	80.6	0.110	83.2	0.114	81.9	1.00	5.69
10.0	2.70	75.9	0.102	77.5	0.105	76.7	1.00	5.33

Table 2

Performance data of Carbon black BP2000-based supercapacitor, measured by both cyclic voltammetry (at a potential scan rate of 20 mV/s), and galvanic charge–discharge curves (at a charging–discharging rate of 20 mA/cm²). The electrolyte is 0.5 M Na₂SO₄ aqueous solution (room temperature). PTFE binder content in the electrode layer: 5 wt%; electrode layer thickness: 100 μm.

Conducting carbon content (wt%)	Active carbon loading, m_{act} mg/cm ²	Specific capacitance from CV, C_s (F/g active carbon)	Electrode geometric capacitance from CV, C' (F/cm ²)	Specific capacitance from GC, C_s (F/g active carbon)	Electrode geometric capacitance from GC, C' (F/cm ²)	Average specific capacitance, C_s (F/g active carbon)	Cell voltage, V_{cell} (V)	Energy density, $E_{\text{spc-act}}$ (Wh/kg active carbon)
5	3.11	80.0	0.126	92.5	0.144	86.3	1.00	6.00
10	3.49	86.1	0.150	88.7	0.155	87.4	1.00	6.07
15	3.00	80.6	0.129	88.2	0.132	84.4	1.00	5.86
20	2.68	75.9	0.107	87.0	0.117	81.5	1.00	5.66

Table 3

Performance data of Carbon black BP2000-based supercapacitor, measured by both cyclic voltammetry (at a potential scan rate of 20 mV/s), and galvanic charge–discharge curves (at a charging–discharging rate of 5 mA/cm²). The electrolyte is 0.5 M Na₂SO₄ aqueous solution (room temperature). Conducting carbon (Super C45) content in the electrode layer: 15 wt%; PTFE binder content: 5 wt%.

Electrode layer thickness, μm	Active carbon loading, m_{act} mg/cm ²	Specific capacitance from CV, C_s (F/g active carbon)	Electrode geometric capacitance from CV, C' (F/cm ²)	Specific capacitance from GC, C_s (F/g active carbon)	Electrode geometric capacitance from GC, C' (F/cm ²)	Average specific capacitance, C_s (F/g active carbon)	Cell voltage, V_{cell} (V)	Energy density, $E_{\text{spc-act}}$ (Wh/kg active carbon)
50	1.46	84.8	0.062	80.7	0.059	82.8	1.00	5.75
100	3.00	86.1	0.129	88.2	0.132	87.2	1.00	6.06
200	5.64	84.1	0.237	87.0	0.245	85.6	1.00	5.94
300	7.94	78.9	0.313	84.8	0.337	81.9	1.00	5.69

Table 4
Performance data of Carbon black BP2000-based supercapacitor, measured by both cyclic voltammetry (at a potential scan rate of 20 mV/s), and galvanic charge–discharge curves (at a charging–discharging rate of 5 mA/cm²). Conducting carbon (Super C45) content in the electrode layer: 15 wt%; PTFE binder content: 5 wt%; electrode thickness: 100 μ m.

Na ₂ SO ₄ concentration, M	Active carbon loading, m_{act} mg/cm ²	Specific capacitance from CV, C_s (F/g active carbon)	Electrode geometric capacitance from CV, C (F/cm ²)	Specific capacitance from GC, C_s (F/g active carbon)	Electrode geometric capacitance from GC, C (F/cm ²)	Average specific capacitance, C_s (F/g active carbon)	Cell voltage, V_{cell} (V)	Energy density, $E_{\text{spec-act}}$ (Wh/kg active carbon)
1	3.12	78.1	0.122	81.1	0.126	79.6	1.00	5.53
0.5	3.00	86.1	0.129	88.2	0.132	87.2	1.00	6.06
0.2	3.02	74.7	0.113	76.9	0.116	75.8	1.00	5.26
0.1	2.80	74.2	0.104	77.4	0.108	75.8	1.00	5.26

the porous electrode layer will become more hydrophobic, leading to a difficulty in both electrolyte penetration and ion mobility inside the matrix structure when aqueous electrolyte is employed. As a result, the electrode performance will be reduced. Therefore, the PTFE content should be optimized. In this paper, several typical PTFE contents were tested with the purpose of electrode layer optimization with respect to the performance.

Table 1 shows the data obtained at various PTFE binder contents in the electrode layer by two methods (cyclic voltammetry (CV) and galvanic charging–discharging curve (GCC)). It can be seen that the specific capacitance differences between those data obtained by CV and those by GCC are insignificant ($\sim 2\%$ difference), suggesting that the data obtained here is reliable.

To see the trend more clearly, both specific capacitance and energy density of the electrode active materials were also plotted as a function of PTFE content, as shown in Fig. 5. It can be seen that a maximum specific capacitance exists (~ 86 – 88 F/g) at a PTFE binder content of 5%. With any further increase in PTFE content, the specific capacitance is decreased. This is mainly because the hydrophobicity of the electrode layer can be increased with increasing PTFE binder content, preventing aqueous electrolyte ions from entering pores in the active carbon particles, leading to less charge accumulation in the double-layer, resulting in reduced specific capacitance. The trade-off between more pore formation and increasing hydrophobicity with increasing PTFE content should give an optimized PTFE content, which is 5 wt% in this paper. In the literature, hydrophilic Nafion[®] ionomer was used as the binder [21], where hydrophobicity did not seem to be an issue in the electrode layer. In the Nafion[®] binder content range from 10 to 30%, no significant influence on specific capacitance could be observed.

3.4. Effect of conducting carbon content

Normally, high surface area activated carbon such as BP2000 gives higher specific capacitance but has a poor electronic conductivity and thus causes a higher cell internal resistance. Conducting carbon has a higher electronic conductivity than activated carbon and is normally used to improve electronic conductivity of the supercapacitor electrode layer. Because the conducting carbon has a low surface area compared to activated carbon, its contribution to electrode capacitance is insignificant. In this paper, both conducting carbons, Super C45 and Super P Li were used in the electrode layers to improve the electronic conductivity of the electrode layer. It was found that the supercapacitor cell with Super C45 electrodes had a lower internal resistance compared to that with Super P Li, and both conducting carbons had a maximum specific capacitance if their contents are at a range of 10–15 wt%. The maximum specific capacitances observed are nearly the same, which is in the range of 88–92 F/g. Table 2 shows the typical supercapacitor performance data when Super C45 is used as the conducting carbon. It can be seen that both methods (CV and GCC) could give the similar values of specific capacitance. Fig. 6 shows both specific capacitance and energy density as a function of conducting carbon content. The maximum values for specific capacitance and energy density could be achieved when the conducting carbon content is in the range of 10–15 wt%.

Drops in both specific capacitance and energy density with increasing conducting carbon content seen in Fig. 6 could be mainly caused by reduced active surface area inside the electrode layer when more conducting carbon with smaller surface area is put into the electrode layer.

3.5. Effect of electrode thickness

Electrode layer thickness directly relates to volumetric capacitance. Thicker electrodes containing more active carbon offer

Fig. 7. Specific capacitance and energy density of carbon BP2000 electrode material as a function of electrode layer thickness. Data from Table 3.

higher volumetric capacitance, as seen in Table 3. Note that the Super C45 conducting carbon and the PTFE binder content used here are 15 wt% and 5 wt%, respectively, which have given optimized performance, as shown in Tables 1 and 2. However, it can be seen from Table 3 that with increasing electrode thickness, the specific capacitance is decreased because the transport of the electrolytic ions into/from active layer becomes more difficult. Therefore, an optimized electrode layer thickness exists. Fig. 7 shows both the specific capacitance and energy density as a function of electrode layer thickness. The maximum values for specific capacitance and energy density could be achieved when the electrode layer thickness is $\sim 100 \mu\text{m}$.

3.6. Effect of electrolyte concentration

In the literature [22], electrolyte concentration showed a strong effect on supercapacitor capacitance. For example, if the electrolyte concentration is high, the ion transport within the electrode layer will be easier, leading to an effective building-up for double layer. However, if the electrolyte concentration is too high, the ion activity may be reduced due to less water hydration, resulting in decreases ion mobility. Therefore, an optimized electrolyte concentration should exist. In general, for aqueous electrolyte based supercapacitors, the most popular electrolyte is aqueous Na_2SO_4 . Therefore, in this work, we employed Na_2SO_4 as a representative system to investigate the electrolyte concentration effect. Table 4 displays the performance data of Carbon black BP2000-based supercapacitor, collected using both cyclic voltammetry and

galvanic charging–discharging curve at different Na_2SO_4 concentrations. In these measurements, the thickness of electrode layer was $100 \mu\text{m}$, the conducting carbon (Super C45) content was 15 wt%, and the PTFE binder content was 5 wt%. This composition of the electrode layer has been identified as the favourite composition as shown in Tables 1–3.

Fig. 8 shows both the specific capacitance and energy density of carbon BP2000 electrode material as a function of Na_2SO_4 electrolyte concentration. It can be clearly seen that both specific capacitance and energy density are the highest when the electrolyte concentration is 0.5 M. When the electrolyte concentration is lower than 0.5 M, both of them are low, which may be due an insufficient number of ions for double-layer building-up. This observation is consistent with the literature [22]. However, when the electrolyte concentration is higher than 0.5 M, both specific capacitance and energy density are reduced, which could be due to reduced ion activity (less water hydration) at higher electrolyte concentrations.

4. Conclusions

In order to optimize supercapacitor performance, the effect of several experimental conditions, such as electrode layer binder content, conducting carbon content, electrode layer thickness, as well as electrolyte concentration, on both the specific capacitance and energy density of an electrode active material (BP2000), were investigated using both cyclic voltammetry and galvanic charging–discharging curves. In electrode layer fabrication, Super C45 carbon was used as the conducting additive, PTFE as the binder, and Na_2SO_4 as the aqueous electrolyte, respectively. It was found that 15 wt% of Super C45 and 5 wt% of PTFE in the electrode layer with a thickness of $100 \mu\text{m}$ could give a better performance in terms of both specific capacitance and energy density. Regarding the effect of electrolyte concentration, it was observed that 0.5 M of Na_2SO_4 could give the best performance within the tested electrolyte concentration range from 0.1 to 1.0 M.

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Fig. 8. Specific capacitance and energy density of carbon BP2000 electrode material as a function of Na_2SO_4 electrolyte concentration. Data from Table 4.

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