CYCLABILITY PERFORMANCE OF ELECTRIC DOUBLE LAYER CAPACITOR WITH GLASS WOOL SEPARATOR

Mohd Ferdaus Mohammad Yaacob & Zulkarnain Ahmad Noorden

Institute of High Voltage and High Current, Faculty of Electrical Engineering,

Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

zulkarnain-an@utm.my

ABSTRACT

The paper presents the cyclability performance of EDLC with glass wool separator in comparison to the conventional cellulose. Two-electrode test cells were constructed by sandwiching two activated carbon electrodes with each separator material. An aqueous electrolyte of sulfuric acid (H_2SO_4) was used as the electrolyte solution. A direct current (DC) characterization test, namely constant current charge-discharge test was carried out on each test cell for up to 300 charge-discharge cycles with constant current of 10 mA. The glass wool-based capacitor performed comparable to the conventional cellulose-based capacitor in terms of its specific capacitance and internal resistance for up to 300 cycles. Thus, by applying glass wool as separator coupled with high concentration aqueous electrolyte may lead to higher rating EDLC at lower cost compared to organic electrolyte- based capacitor.

1. INTRODUCTION

Electric double layer capacitor (EDLC) is one of promising supercapacitors that capable of storing high energy with small form factor. It stores charges electrostatically onto its high surface area carbon electrodes. Since the charge storage mechanism does not involve any chemical reaction, - oxidation and reduction processes - EDLC possesses higher charge-discharge cyclability compared to pseudo-capacitor and lithium-ion capacitor. Needless to say, many researchers are now looking to improve the EDLC performances in terms of power and energy densities as well as cycle ability [Kado, et al (2014), Kim (2015), Lui, et al. (2011), Senthilkumar et al. (2013), Tonurist, et al. (2012), Weng, et al. (2008) and Zhang, et al. (2015)].

Current major disadvantage of commercial supercapacitor is lower specific stored energy typically below 10 Wh/kg compared to lowest specific stored energy for battery, which is up to 40 Wh/kg [Obreja (2007)]. For EDLC, the energy capacity is dominated by its capacitance value, which is significantly depends on the surface area of the active electrodes, properties of the electrolyte ions, and the electrolyte concentration [Inagaki, et al. (2010)]. The electrolyte may be in aqueous solution, polymer gel, or organic electrolyte [Lewandowski, et al. (2010)].

Previous studies proved that the specific capacitance of EDLC increased with the increment of electrolyte concentration [Weng, et al. (2008) and Soneda, et al. (2004)]. Aqueous solution - a low cost electrolyte [Kotz (1999)], such as aqueous sulfuric acid, (H_2SO_4) offers higher power density of EDLC compared to organic electrolyte [Lewandowski, et al. (2010)].

One way to achieve higher performance EDLC is by utilizing high concentration aqueous electrolyte coupled with corrosive resistance separator material. By applying high concentration of H_2SO_4, high energy density can be achieved with much lower cost compared to organic electrolyte.

EDLC with corrosive resistance separator, glass wool has been reported previously since most of conventional separators such as cellulose are incapable to resist high concentration of aqueous electrolyte [Noorden, et al (2014)]. Conventional cellulose separator for EDLC tends to degrade with high concentration of aqueous electrolyte. It is found that glass wool-based EDLC has outperformed conventional EDLC with cellulose separator in average of 38% and 9% difference of power and energy densities, respectively [Noorden (2013 & 2014)]. Nonetheless, currently there is no report concerning on its cyclability performance.

This paper evaluates the cyclability of the corrosive resistance material, glass wool as separator for EDLC application under constant current charge-discharge test.

The cycle performances of glass wool and conventional separator, cellulose paper were compared and investigated through two-electrode capacitor system with 1 mol/dm^3 of aqueous H_2SO_4 as the electrolyte.

2. EXPERIMENT

2.1 Electrolyte Preparation

An aqueous H_2SO_4 with concentration of 1 mol/dm^3 was prepared by using 95-97% concentrated H_2SO_4 (QreC, QREC (Asia) Sdn. Bhd., Malaysia) through dilution process at room temperature. A 3.33 ml of the concentrated H_2SO_4 was added to 30 ml deionized water and then the solution was adjusted by adding more de-ionized water up to 60 ml. The volume of solution was calculated based on following equation:

$$M_1 V_1 = M_2 V_2 \tag{1}$$

where M_1 (mol/dm^3) is molarities of concentrated H_2SO_4, V_1 (ml) represents volume of concentrated H_2SO_4, M_2 (mol/dm^3) is the desired concentration of diluted H_2SO_4, and V_2 (ml) is the desired volume for diluted H_2SO_4. Above equation may be used with any concentration units as long as the units are the same of each side.

2.2 Test Cell Construction

Glass wool (CMAX, CNBM International Corporation, China) and cellulose paper (TF4030, Nippon Kodoshi Corporation, Japan) were cut in circle shape and weighed before moistened with the 1 mol/dm^3 H_2SO_4. The measurements were recorded in Table 1. As shown in Fig. 1, each electrolyte-containing separator was inserted between two symmetrical activated carbon sheets (Nippon Valqua Industries Ltd., Tokyo, Japan) as the electrodes. The two-electrode test cell system was then manually connected to Gamry Instrument Interface 1000 and the measurements were taken under room temperature.

3. RESULTS AND DISCUSSION

The voltage responses of the tested capacitor at 10 mA charge-discharge current are shown in Fig. 2. The linear voltage response confirmed that the tested capacitors behave similar to typical EDLC without any chemical reaction involved [Noorden, et al. (2014)]. The test cell was set up to charge up to 1 V, which is the maximum voltage window for aqueous electrolyte [Kotz (1999)]. Unlike ideal capacitor, both glass wool and cellulose test cell capacitors encounter voltage drop during charge-discharge transition process. This is due to the internal resistance, namely as equivalent series resistance, R_{int} (Ω) and can be calculated with following relationship;

Table 1 Measurement of carbon electrodes for both separators before moistened with 1 mol/dm^3 H_2SO_4.

	Glass wool		Cellulose	
	Diameter (mm)	Mass (mg)	Diameter (mm)	Mass (mg)
Carbon 1	19	35.6	19	35.2
Carbon 2	19	35.3	19	35.1
Separator	36.9	141.4	36.9	14.2

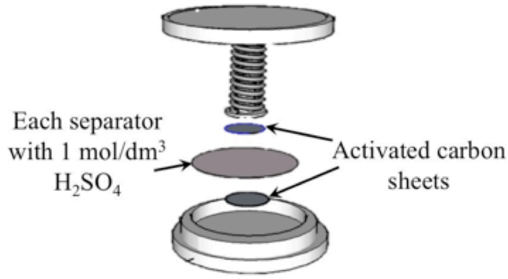

Fig. 1 Two- electrode test cell configuration.

$$R_{int} = V_{drop} / \Delta I \tag{2}$$

where V_{drop}, (V) is the voltage drop and ΔI (A) is the current change at instance of charge-discharge transition.

The R_{int} for both tested capacitors at 100th cycle are approximately 12 Ω. The value of R_{int} against cycle number was plotted in Fig. 3. The graph shows that R_{int} for glass wool is slightly higher compared to cellulose paper after 50 cycles. However, no significant difference observed up to 300-cycle charge-discharge. At 300th cycle, the R_{int} of 11.8 Ω and 11.3 Ω were recorded for glass wool and cellulose-based capacitors, respectively. The finding suggests that the tested capacitor with glass wool separator possesses a comparable performance to the one with cellulose in terms of its internal resistance for up to 300 cycles.

The capacitance, C (F) of the test cell value was computed based on the following equation;

$$C = Q/V \tag{3}$$

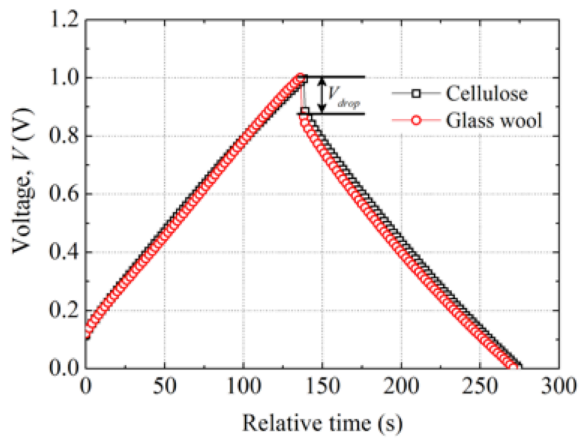

Fig. 2. Voltage responses for cyclic charge-discharge with 10 mA constant current at 100th cycle for tested capacitor.

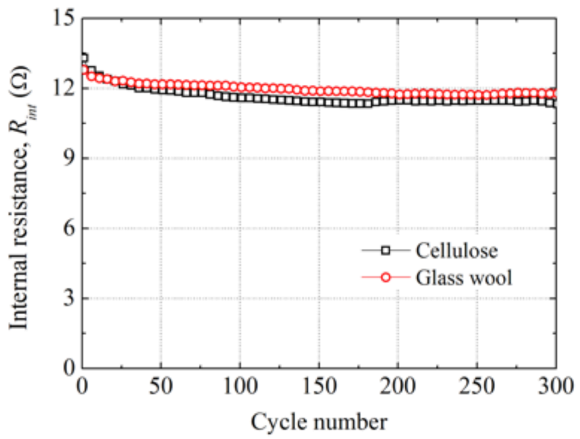

Fig. 3. Internal resistance of the tested capacitors for 300 cycles.

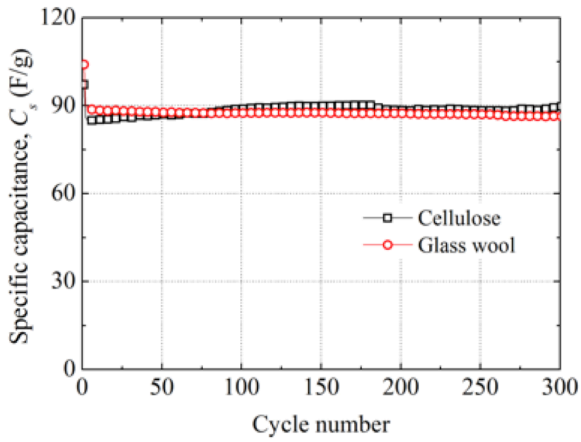

Fig. 4. Specific capacitance of the tested capacitors for 300 cycles.

where Q (C) is the charge taken from current measurement and V (V) denotes as the voltage response.

The overall of test cell capacitance, C_{cell} can be calculated based on following equation;

$$C_{cell} = (C_c + C_d)/2 \tag{4}$$

where C_c *and* C_d are capacitance values during charging and discharging process respectively. The test cell capacitance, C_{cell} (F) was then converted to specific capacitance, C_s (F/g) using following equation;

$$C_s = C_{cell} * 4/m \tag{5}$$

where m (g) represents the total weight of activated carbon electrodes.

The specific capacitance C_s was then plotted against cycle number as shown in Fig. 4. Referring to the figure, capacitance losses for glass wool-based capacitor is 2.4%. The reduction is in contradiction to cellulose separator where the capacitance increases about 5.4%. However, in terms of the specific capacitance, the cyclability performance of the glass wool-based capacitor seems more stable for up to 300 cycles compared to the capacitor with cellulose separator, which resulted a fluctuating capacitance throughout the cyclability test. The fluctuated capacitances graph as shown in Fig. 4 may due to the effect on the ionic charge compensation rate between positive and negative charges micro porous carbon electrodes soaked into electrolyte solution. The molar conductivity of ions in separator is extremely important to ensure the stability of charge-discharge activities [Liivand, et al. (2015)]. The C_s for cellulose-based capacitor is slightly higher (89.7 F/g) compared to the one with glass wool separator (86.3 F/g). Nevertheless, the finding also suggests that in terms of specific capacitance, the glass wool-based capacitor has performed comparably to the one with cellulose separator.

4. CONCLUSION

Based on the cyclic charge-discharge process, the corrosive resistance material, glass wool separator performance behaves comparable to the conventional cellulose separator under 1 mol/dm^3 H_2SO_4 in term of specific capacitance, C_s and internal resistance, $R_{int,}$ This findings indicate that glass wool is a promising separator to replace cellulose paper in order to utilize high concentrated electrolytes in EDLC construction. The extended studies on such electrolytes in EDLC with glass wool as the separator need to be conducted.

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Mohd Ferdaus Mohammad Yaacob received the B.Eng. degree in electrical-electronics engineering from Universiti Teknology Malaysia (UTM), in 2011. He is presently pursuing the master's degree under Department of Electrical Engineering. His research interests include energy storage system and technology.

Zulkarnain Ahmad Noorden received the Ph.D. degree in electrical engineering from Shibaura Institute of Technology Japan in 2013. He is currently works at Universiti Teknologi Malaysia (UTM) as a senior lecturer. His research interests include energy storage system and power equipment diagnosis.