

Preparation of activated carbon electrodes from orange peel biomass with various separator materials for supercapacitor applications

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ABSTRACT

Activated carbon electrodes from orange peel biomass materials for supercapacitor applications with variety of type separator have been prepared. Activated carbon was prepared by pyrolysis process at 800° under N₂ gas environment. Electrochemical characterization was tested on a variety of separators, i.e., JR-800-W (using Whatman paper number 40), JR-800-E (using eggshell membrane, and JR-800-O (using orange fruit membrane). The results of chemical measurement for the cyclic voltammetry method on the three samples are capacitance values of 191.82 F/g on JR-800-W, 115.08 F/g on JR-800-E, and 94.17 F/g on JR-800-O. The capacitance value in the galvanostatic charge-discharge method are 174.24 F/g with IR drop of 0.067 for sample JR-800-W, 133.22 F/g with IR drop of 0.14 for sample JR-800-E, and 116.8 F/g with IR drop of 0.36 for sample JR-800-O. Whatman paper separators produce good electrochemical properties, indicating the use of separators can affect the performance of activated carbon electrodes for supercapacitor applications.

Keywords: Activated carbon electrode; orange peel; separator; supercapacitor

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INTRODUCTION

According to the International Energy Agency (IEA), worldwide electrical energy consumption has increased significantly in the past five years. Global electrical energy consumption reached 22,000 TWh in 2019, and will increase to 28,000 TWh in 2023. They predict that the world's Electricity consumption will grow 3.4% through 2026. Almost all electrical energy comes from fossil fuels [1].

The topic of energy storage technology is increasingly becoming a concern discussed by various groups. Energy storage media such as batteries and capacitors are considered effective as low-voltage storage solutions. However, batteries and capacitors are not yet the best solution for maximum energy storage. The power density is not too high and the charging time is relatively long which is a shortcoming of the battery, so it is necessary to innovate to create other alternative energy storage in large quantities and can be used for a long time. Supercapacitors offer significant advantages and have been widely applied in modern

society, especially in managing renewable energy sources. One of the promising applications is wearable devices, electric vehicles, and stationary energy storage systems. Supercapacitors are becoming significant energy conversion devices due to their extremely fast charge/discharge rate, environmentally friendly, durable, independent of time and weather conditions, and high specific power exceeding batteries [2]. One aspect that affects the effectiveness of supercapacitors is the use of electrode materials. Carbon is one type of material used for making electrodes because of its high specific surface area. Various types of carbon are used in making electrodes, one of which is activated carbon. Activated carbon also known as activated charcoal is a type of graphite that is rough and not perfectly structured. Activated carbon is characterized by a wide spectrum of pores of various sizes [3].

Conventional separator materials used in supercapacitors such as rubber, plastic films, aquagels, resorcinol formaldehyde polymers,

and polyolefins have been employed primarily to prevent electronic conduction between the electrodes. However, these materials often suffer from long-term issues such as drying, structural collapse, or low ionic conductivity. Therefore, there remains a critical need for separator materials that are highly porous to facilitate efficient ionic transport while simultaneously acting as effective electronic insulators between opposing electrodes. The type of separator material that is widely used at present is the nafiion membrane which, although efficient for the role of supercapacitor separator, is very expensive and has limited availability [4].

Orange peel waste is one of the most common wastes found in the environment. Mueller (2017) [5] reported that in 2014 citrus production worldwide reached 68 million tons, most of which was used for the juice industry which produced 3.8 million tons of orange peel waste per year. Sweet orange peel contains 6%-19.801% lignin, 46-69% cellulose, and 6% hemicellulose [6]. Sweet orange peel has abundant cellulose compared to rice merang 33%-43% [7], pine seeds 44% [8], coconut fiber 43,44% [9] which indicates an abundant carbon source so that it has the potential to be used in making activated carbon if given the right treatment.

The main objective of this work was to analyze the effect of different type of separator materials on the performance of supercapacitor activated carbon electrodes using Whatman paper, eggshell membrane, and orange fruit membrane.

MATERIALS AND METHOD

Activated Carbon Preparation

Orange peel waste was obtained and collected from Pekanbaru City, Riau province. The processing of orange peel waste includes drying under sunlight and using an electric oven at 110°C for 2 hours, pre-carbonization at 200°C for 2 hours. Sample was activated using ZnCl₂ 0.5 M. The pyrolysis process was carried

out under N₂ gas pressure at temperatures of 800°C.

Preparation of Separator

The separator materials selected were Whatman paper grade 40, eggshell membrane, and orange fruit membrane. The three separator materials were soaked using sulfuric acid (H₂SO₄) with a concentration of 1 M. Samples are coded JR-800-W for whatman paper separator, code JR-800-E for eggshell membrane separator, and code JR-800-O for orange fruit membrane separator.

Physical - Electrochemical Characterization

Physical characterization was performed by measuring electrode density by measuring mass and volume, while electrochemical performance was evaluated using cyclic voltammetry (CV) at scan rates of 1, 2, 5 mV/s. Galvanostatic charge-discharge (GCD) to determine specific capacitance at a current density of 1 2, 5 A/g with a maximum potential of 1 V, and internal resistance.

RESULTS AND DISCUSSION

Physical Property Analysis

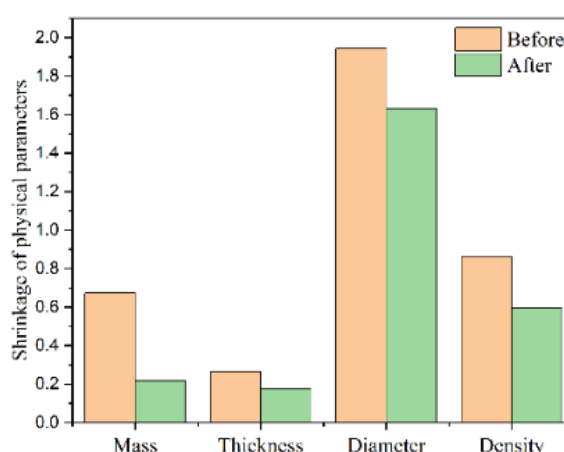


Figure 1. Electrode density before and after pyrolysis process.

Figure 1 shows the electrode density before and after the pyrolysis process. The average density is calculated using the Equation (1):

$$p = \frac{m}{v} \quad (1)$$

where, m is the average mass, and v is the average volume of the electrode. The resulting decrease in electrode density is 0.268388 g/cm^2 with a percentage of 31.125%, causing more volatile compounds to be lost, thus reducing the density of the sample. The decrease in density causes the carbon pore walls to collapse, resulting in high porosity. Pyrolysis process

depends on temperature, pressure, and the composition contained in the biomass to produce an increase in the number of pores contained. Composition occurs at different temperature levels. Cellulose, hemicellulose compounds are decomposed at $200^\circ\text{C} - 350^\circ\text{C}$ which evaporates volatile compounds, and lignin can be decomposed at temperatures above 500°C which produces products such as tar and activated charcoal [10].

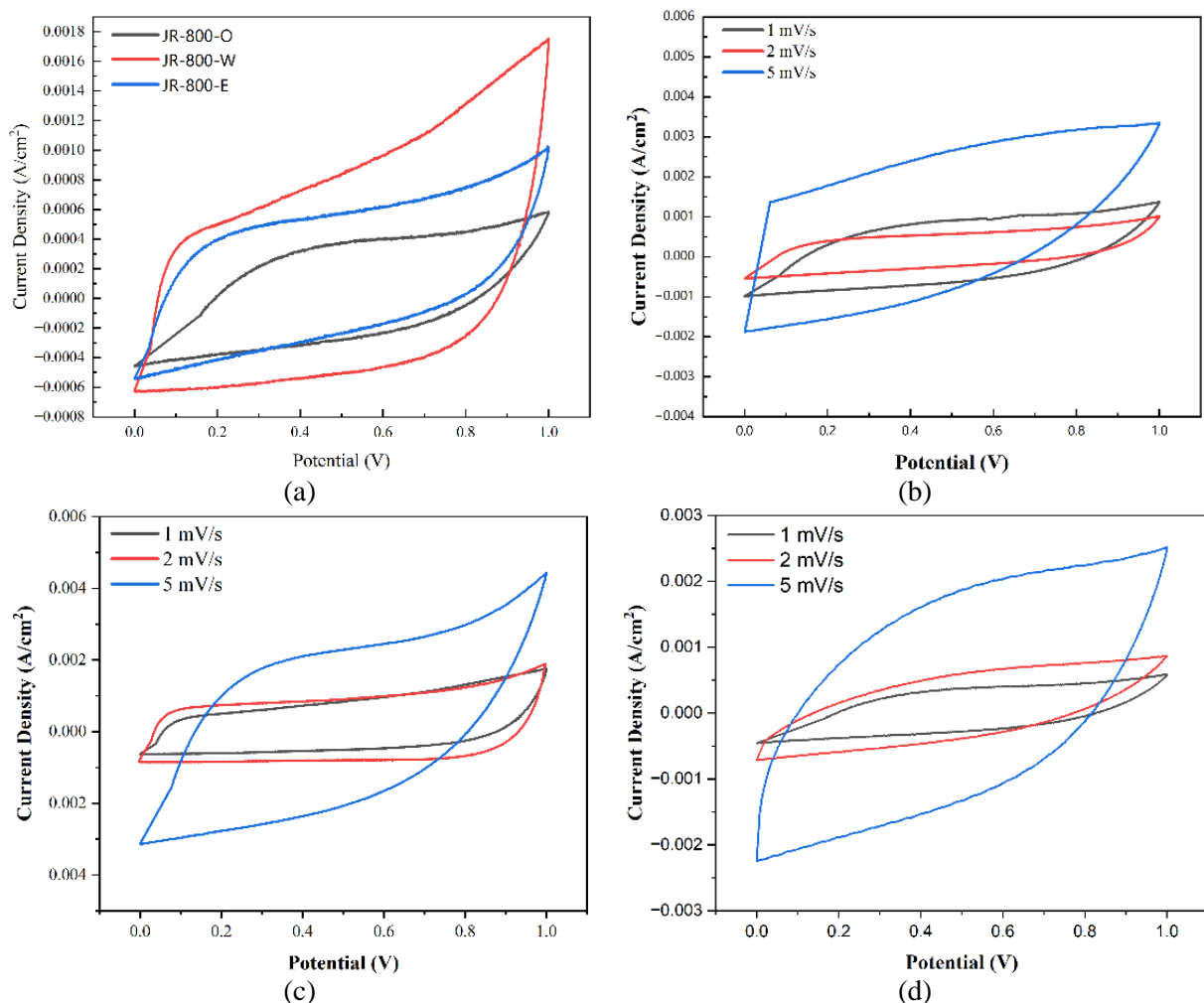


Figure 2. CV curve of (a), various separator, (b), JR-800-O (c), JR-800-W (d), JR-800-E at various scan rate.

Electrochemical Properties

CV (Cyclic Voltammetry) measurement results from orange peel carbon electrodes on variations of JR-800-O, JR-800-W, and JR-800-E separator materials. The specific capacitance results of the three variations are highest in JR-

800-W at 191.82 F/g , followed by JR-800-E at 115.08 F/g , and JR-800-O at 94.17 F/g . This is reinforced by Figure 2 (a) which shows that the JR-800-W curve area is larger, indicating that more ions participate in the formation of the electric double layer [11]. Whatman paper separators have a uniform microporous

structure and excellent electrolyte absorption, which allows ions in the electrolyte to diffuse quickly and efficiently towards the electrode surface. This structure supports optimal formation of an electrical double layer, resulting in high capacitance [6]. The eggshell membrane is composed of interwoven fibrous protein layers (primarily collagen and keratin), which exhibit limited ionic conductivity and uneven surface morphology. These factors hinder full electrolyte infiltration and create tortuous ion pathways, thereby reducing effective ion transport and charge storage capability. Nevertheless, its partial permeability allows better performance than dense plant-based membranes, supporting its viability as a low-cost bioseparator [4]. The inferior

performance of this separator is attributed to its dense and heterogeneous structure, characterized by compact cellulose-lignin cell wall layers with low porosity and poor electrolyte affinity. These features impede electrolyte absorption and slow down ion migration, which not only limits the active surface area for electric double layer formation but also introduces significant internal resistance. Moreover, the non-uniform swelling behavior upon contact with electrolyte contributes to mechanical instability and inconsistent ionic pathways across the electrode-separator interface [6]. The JR-800-W remains the best performer showing good electrochemical, stability and performance at various scan rates [12].

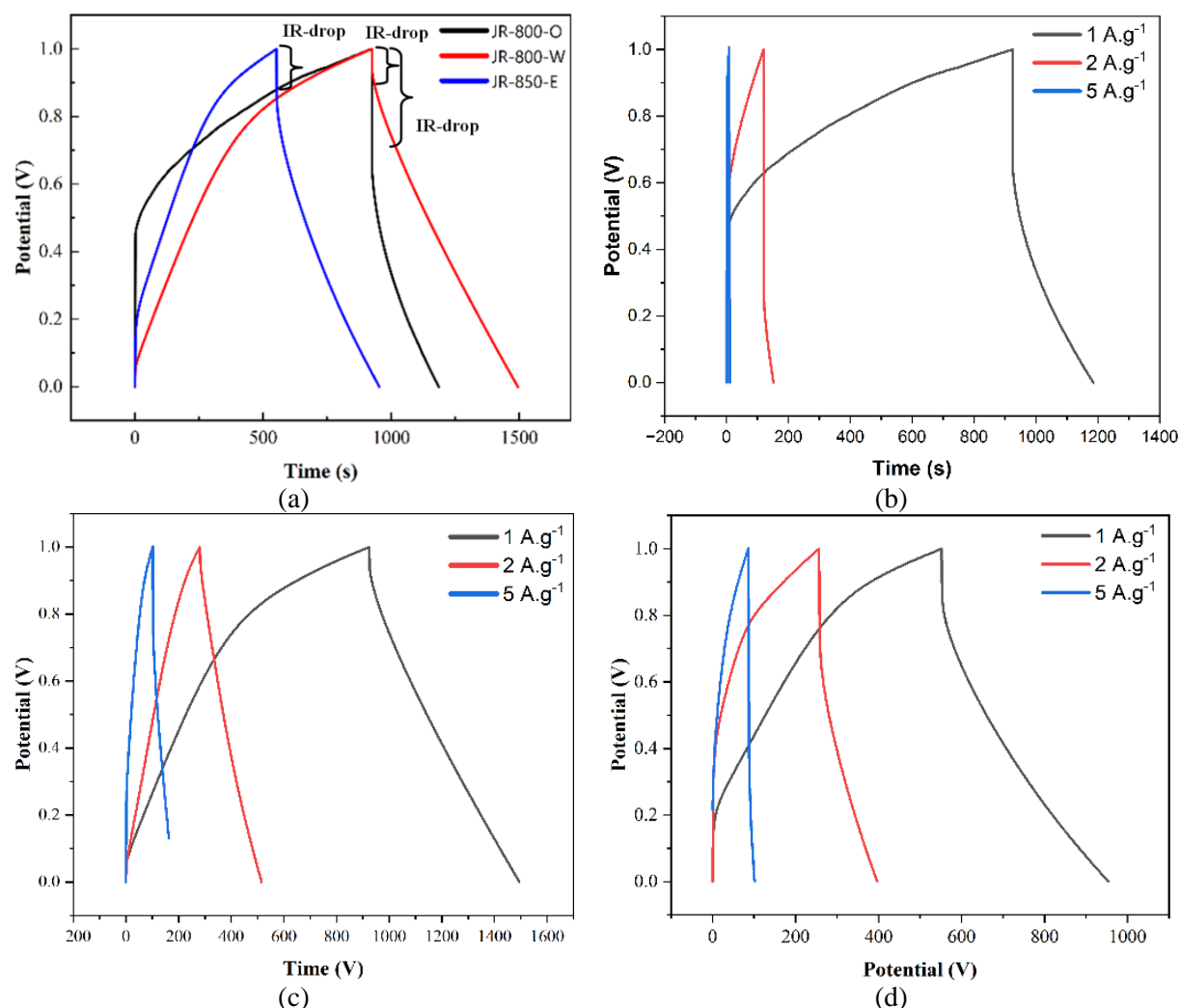


Figure 3. GCD curve of (a), various separator, (b), JR-800-O (c), JR-800-W (d), JR-800-E at various current densities.

The GCD curve Figure 3 (a) shows a symmetrical triangular shape indicating high reversibility of the carbon electrode as an EDLC. As a result, JR-800-W again showed the highest value. IR drop is seen to be the smallest in JR-800-W at $0.067\ \Omega$ with a specific capacitance value of $174.24\ \text{F/g}$ followed by JR-800-E at $0.14\ \Omega$ with a specific capacitance value of $133.22\ \text{F/g}$, and JR-800-O at $0.36\ \Omega$ with a specific capacitance value of $116.8\ \text{F/g}$, indicating lower internal resistance in JR-800-W and better energy efficiency. The low IR drop in the Whatman separator is due to the uniform microstructure, making the diffusion of electrolyte ions easier and more efficient. Eggshell membranes have an inhomogeneous structure due to the uneven structure of the collagen layer inhibiting ion diffusion between surfaces resulting in higher internal resistance than whatman separators. Low porosity is possessed by the orange membrane separator, resulting in the highest internal resistance. This is in line with previous findings. Figure 3 (b), (c), (d) shows that an increase in current density leads to a decrease in specific capacitance values for all samples. This is because at high current densities, electrolyte ions have less time to diffuse into the electrode pores [13], however, JR-800-W retains relatively high values, supporting previous findings.

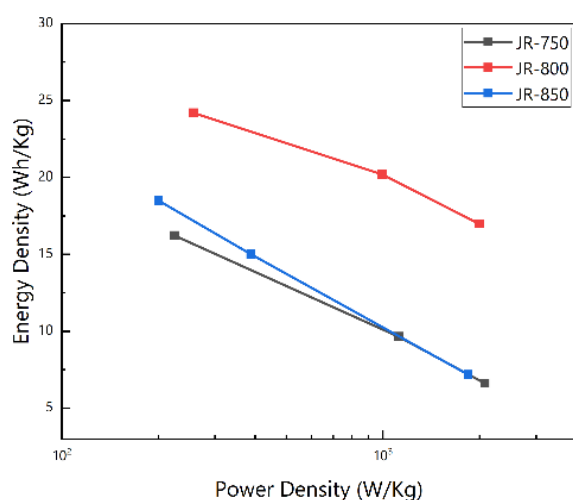


Figure 4. Ragone plot.

Figure 4 show the Ragone plot of three separator variations. This reflect the optimal ion

accessibility and minimal internal resistance offered by the uniform microstructure and excellent wettability of the Whatman paper separator variations. This reflect the optimal ion This finding aligns with previous reports that emphasize the role of high-porosity, thin separators in improving ion transport kinetics and minimizing IR drop, thus enhancing both energy and power output simultaneously [11, 4].

In contrast, the JR-800-E and JR-800-O samples exhibited a significant trade-off between energy and power densities. The eggshell membrane (JR-800-E), while biologically derived and cost-effective, exhibited a moderately lower E_{sp} and P_{sp} . This reduction is attributed to the membrane's irregular fibrous morphology and semi-permeable protein layers that introduce tortuosity in ion pathways and increase ionic resistance [4]. Similarly, the JR-800-O separator produced the lowest performance, with an estimated likely due to its dense cellulose-lignin structure, poor electrolyte uptake, and limited ionic mobility.

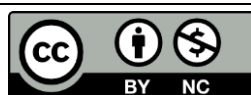
CONCLUSION

This study demonstrated that the type of separator material significantly influences the electrochemical performance of activated carbon electrodes derived from orange peel biomass for supercapacitor applications. Among the three separators tested Whatman filter paper no. 40 (JR-800-W), eggshell membrane (JR-800-E), and orange fruit membrane (JR-800-O) the JR-800-W sample consistently exhibited the highest specific capacitance in both CV ($191.82\ \text{F/g}$) and GCD ($174.24\ \text{F/g}$) analyses. This superior performance is attributed to the uniform microporous structure and excellent electrolyte absorption of Whatman paper, which promotes faster ion diffusion and more efficient electric double-layer formation. The eggshell membrane separator (JR-800-E) showed moderate performance, with sufficient ionic mobility but slightly higher internal resistance.

Meanwhile, the orange fruit membrane (JR-800-O) demonstrated the lowest performance due to its dense, non-uniform structure and limited electrolyte uptake, resulting in reduced ion transport and higher IR drop. These findings affirm that choosing a separator with high porosity, good wettability, and mechanical stability is critical to optimizing the energy storage efficiency and cycling stability of supercapacitor devices.

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