

## Harnessing multi-doping porous carbon from *Musa paradisiaca* L. peel waste for solid-state supercapacitors

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### ABSTRACT

The demand for low-cost carbons with multi-doping in supercapacitors has led to a significant focus on utilizing biomass waste to produce activated carbons. The research successfully utilized *Musa paradisiaca* L. (MPL) peel as a porous carbon for solid-state supercapacitor. The process involved collecting banana peel waste, drying the peels using sunlight, pre-carbonization using a furnace, pH neutralization, drying, crushing carbon particles, and ensuring uniform particle size. Different concentrations of the catalytic ZnCl<sub>2</sub> solution (300, 500, and 700 mmol/g) were selected to optimize physical and electrochemical properties. The resulting chemically activated MPL carbon powder was evaluated using SEM-EDS, XRD, and BET. MPL activated carbon with a 500 mmol/g solution of ZnCl<sub>2</sub> was found to have optimal physical properties with a carbon percentage of 81.65%, oxygen 17.39%, phosphorus 0.42%, and boron 0.52%. Electrochemical properties were evaluated using dual-electrode system was exhibited the highest specific capacitance of 67 F/g. These findings demonstrate the potential of MPL peel waste as a high quality electrode for supercapacitor next-generation.

**Keywords:** Carbon; electrode materials; *Musa paradisiaca* L. peel; self-doped; supercapacitor

Received 06-09-2023 | Revised 10-10-2023 | Accepted 29-10-2024 | Published 30-11-2024

### INTRODUCTION

Electrical energy has become a fundamental necessity for people worldwide and has significantly impacted almost every aspect of life. In Indonesia, the reliance on fossil fuels like oil, coal, and natural gas for electrical energy has led to air pollution, affecting the quality of life and public health [1]. The country's energy demand continues to rise due to economic activities in the industrial, transportation, commercial, and household sectors. This escalating energy requirement has prompted the exploration of various alternative energy sources to address the national energy crisis. Renewable energy sources such as solar and wind can generate electrical energy, but

their effective use depends on efficient energy storage [2].

Supercapacitors are promising electrical energy storage technologies for the future. Supercapacitors are widely used in various applications such as in vehicles, trains, electronics, and in telecommunications. This technology offers energy storage with high power density and a long life cycle [3]. This is because supercapacitors store energy electrostatically through the absorption of electrolyte ions at the electrolyte electrode interface [4]. This non-faradaic process produces high power density but low energy density. Furthermore, its long life cycle occurs because supercapacitors do not experience significant degradation as occurs in

conventional batteries [5]. Among the components of supercapacitors, the selection of electrode materials is very important in improving their performance. Carbon materials have been widely studied as supercapacitor electrode materials because they have superior electrical conductivity, good chemical stability, and high surface area. However, carbon materials mostly come from fossil fuels whose application in the future industrial sector is limited due to their high cost, non-renewable sources, complicated synthesis, and environmental pollution. Thus, it is important to find carbon materials that are low-cost, easy to synthesize, and environmentally friendly.

Biomass-based carbon materials have attracted much attention due to their low cost, sustainability, and wide availability in nature [6]. Several biomass materials that have been utilized such as coconut shells have been successfully used as carbon sources with high carbon content and have O, N, and B doping [7]. Furthermore, hemp biomass has been successfully used as activated carbon material through high-temperature pyrolysis and chemical activation using KOH [8]. Garlic peel waste has been successfully used as porous activated carbon with the addition of melamine dopant and  $\text{KHCO}_3$  activator selected as activator material [9]. In addition, other biomass materials that have also been successfully used as activated carbon are soybean straw [10], hazelnuts [11], and bamboo [12]. However, the methods used in these studies limit their large-scale application due to their complicated synthesis, longer time, and high cost [13]. Therefore, an easier, simpler, lower cost, and more time-saving method is needed.

On the other hand, bananas are one of Indonesia's plantation products that are produced in large quantities every year. Based on data from the Central Statistics Agency, another part of the banana that can also be used is the peel which contains 14.4% cellulose. In recent years, banana peels have been widely studied for use as absorbent materials (activated carbon). In more detail, *Musa paradisiaca* L.

(MPL) peels contain 43.7% carbon elements consisting of cellulose, hemicellulose, chlorophyll pigments, and pectin substances so that they support the production of activated carbon as a supercapacitor electrode material. These components support the potential of MPL peels to be used as activated carbon with a high degree of crystallinity so that the activated carbon obtained can be applied to supercapacitors. MPL peels are synthesized with easy and cost-effective preparation, starting with the drying process, pre-carbonization using a furnace, chemical activation, and high-temperature pyrolysis. Chemical activation was carried out using a  $\text{ZnCl}_2$  activator with varying concentrations of 300, 500, and 700 mmol/g. The electrochemical performance of the MPL peels-activated carbon produced was tested using a two-electrode configuration, showing a specific capacitance of 67.14 F/g, with an energy density of 9.325 Wh/kg and a power density of 33.60 W/kg. The results of this study indicate that MPL peels waste can be used as an activated carbon material for supercapacitor electrodes.

## MATERIALS AND METHODS

The MPL peel was cut into small pieces and then dried in the sun to reduce its water content. Furthermore, the pre-carbonization process was carried out on 5 kg of MPL peel samples in one burning using a furnace. The samples that had gone through the pre-carbonization process were then soaked in distilled water (equivalent) until  $\text{pH} = 7$ , then the samples were dried in the sun. Furthermore, the sample refinement process was carried out using a mortar and ball milling tool. The sample was first ground until smooth using a mortar, then the sample was put into a vial for the ball milling process for approximately 24 hours. The sample was sieved using a sieve measuring  $\leq 60 \mu\text{m}$  to obtain uniform results.

Next, chemical activation was carried out using  $\text{ZnCl}_2$  activator. The ratio of activator and carbon is 1 : 5, namely 30 grams of sample was inserted, then 150 ml of distilled water was

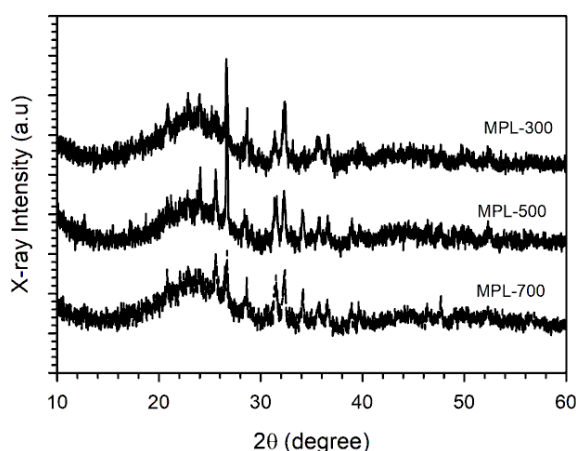
used. The first stage in this chemical activation is to dissolve the  $\text{ZnCl}_2$  activator into 150 ml of distilled water using a hot plate stirrer for 1 hour at  $80^\circ\text{C}$ . The MPL peel carbon sample was inserted into the activator mixture and stirred using a magnetic stirrer for 2 hours at  $80^\circ\text{C}$ , then dried using an oven. The dried sample was ground until smooth and then sieved using a  $60\ \mu\text{m}$  sieve. The MPL peel carbon sample was pyrolyzed at high temperature using a furnace. Furthermore, the sample was neutralized again so that the remaining combustion powder did not mix with the activated carbon sample.

The degree of crystallinity of MPL was evaluated using X-ray diffraction (XRD) in the angle range ( $2\theta$ )  $10^\circ - 60^\circ$  with a Shimadzu type MAXima\_X XRD 7000. The surface morphology of MPL was observed using a scanning electron microscope (SEM) at magnifications of  $5000\times$  and  $15000\times$  and the elemental composition of MPL was evaluated through energy dispersive spectroscopy (EDS) using a Jeol JSM-IT200 instrument. The porosity properties of MPL were evaluated using a Quantachrome Nova 4200e instrument to determine the specific surface area and pore size distribution through the Brunauer–Emmett–Teller (BET) method. The electrochemical performance of the supercapacitor cell was evaluated using cyclic voltammetry (CV) and galvanostatic charge discharge (GCD) in a two-electrode system. Previously, the MPL-activated carbon powder sample was molded using a hydraulic press in the form of a monolithic coin and additional materials. The molded sample was then

polished with a diameter and thickness of 8 mm and 0.2 mm, respectively, and immersed in 1 M  $\text{H}_2\text{SO}_4$  electrolyte. The CV method at a maximum voltage window of up to 1 V used a CV UR Rad-ER 5841 physics instrument. The GCD method was carried out at a current density of 1 A/g using a CD UR Red-ER 2018.

## RESULTS AND DISCUSSION

The samples of activated carbon from banana peel based on the influence of different concentrations of activators MPL-300, MPL-500, and MPL-700 showed XRD patterns in the form of two gentle peaks and several sharp peaks [14]. The MPL-300, MPL-500, and MPL-700 samples had peaks related to each scattering plane, where on the (002) and (100) planes in the range of  $22.987^\circ$ ,  $23.241^\circ$ ,  $23.195^\circ$  and  $44.968^\circ$ ,  $44.734^\circ$ ,  $43.130^\circ$ . The reflection angles on the hkl 002 and 100 planes indicate that the activated carbon samples based on banana peel have an amorphous structure.



**Figure 1.** XRD pattern of MPL peels sample.

**Table 1.** Carbon lattice parameters of MPL peels.

Sample code	$2\theta_{002}$ ( $^\circ$ )	$2\theta_{100}$ ( $^\circ$ )	$d_{002}$ ( $\text{\AA}$ )	$d_{100}$ ( $\text{\AA}$ )	$L_c$ ( $\text{\AA}$ )	$L_a$ ( $\text{\AA}$ )
MPL-300	22.987	44.968	3.865	2.014	8.271	6.799
MPL-500	23.241	44.734	3.824	2.024	8.650	22.766
MPL-700	23.195	43.130	3.831	2.095	10.933	2.320

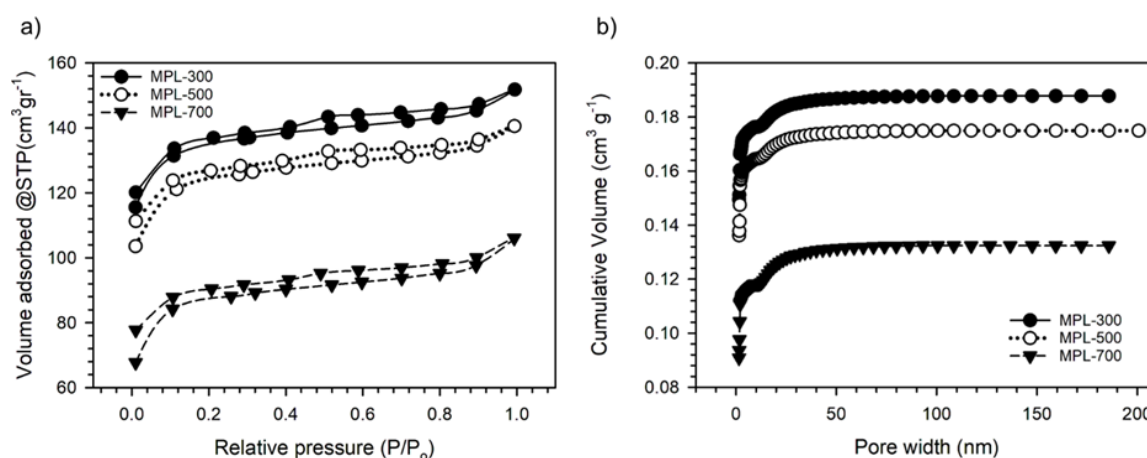
The data presented in Table 1 shows the lattice parameters that provide information on the shift of the  $2\theta$  angle that causes changes in the value of the microcrystalline layer in each

sample. Giving the concentration of  $\text{ZnCl}_2$  to the MPL peel sample from 300 to 700 mmol/g can increase the height value of the microcrystalline layer ( $L_c$ ) from 8.271 to

10.933 Å. Then, a small  $L_c$  value indicates a larger specific surface area [15]. More details are discussed in the  $N_2$  gas absorption analysis.

The pore characteristics of MPL were investigated through nitrogen adsorption/desorption measurements shown in Figures 2 (a) and (b). The isotherm curves between relative pressure ( $P/P_0$ ) and STP volume ( $cm^3/g$ ) where the relationship is in accordance with the IV isotherm based on the IUPAC classification. The hysteresis loops show varying shapes related to capillary

condensation in the mesoporous layer depending on the concentration ratio of the activating agent and the synthesis form used [16]. The combination of macropores, mesopores, and micropores can support electrolytes to shorten the ion diffusion distance and facilitate rapid ion transfer. This type of curve forms micropores and mesopores which simultaneously contribute to the enhancement of the electrochemical double layer capacitance (EDLC) and improve the accessibility of high power density of supercapacitor devices.



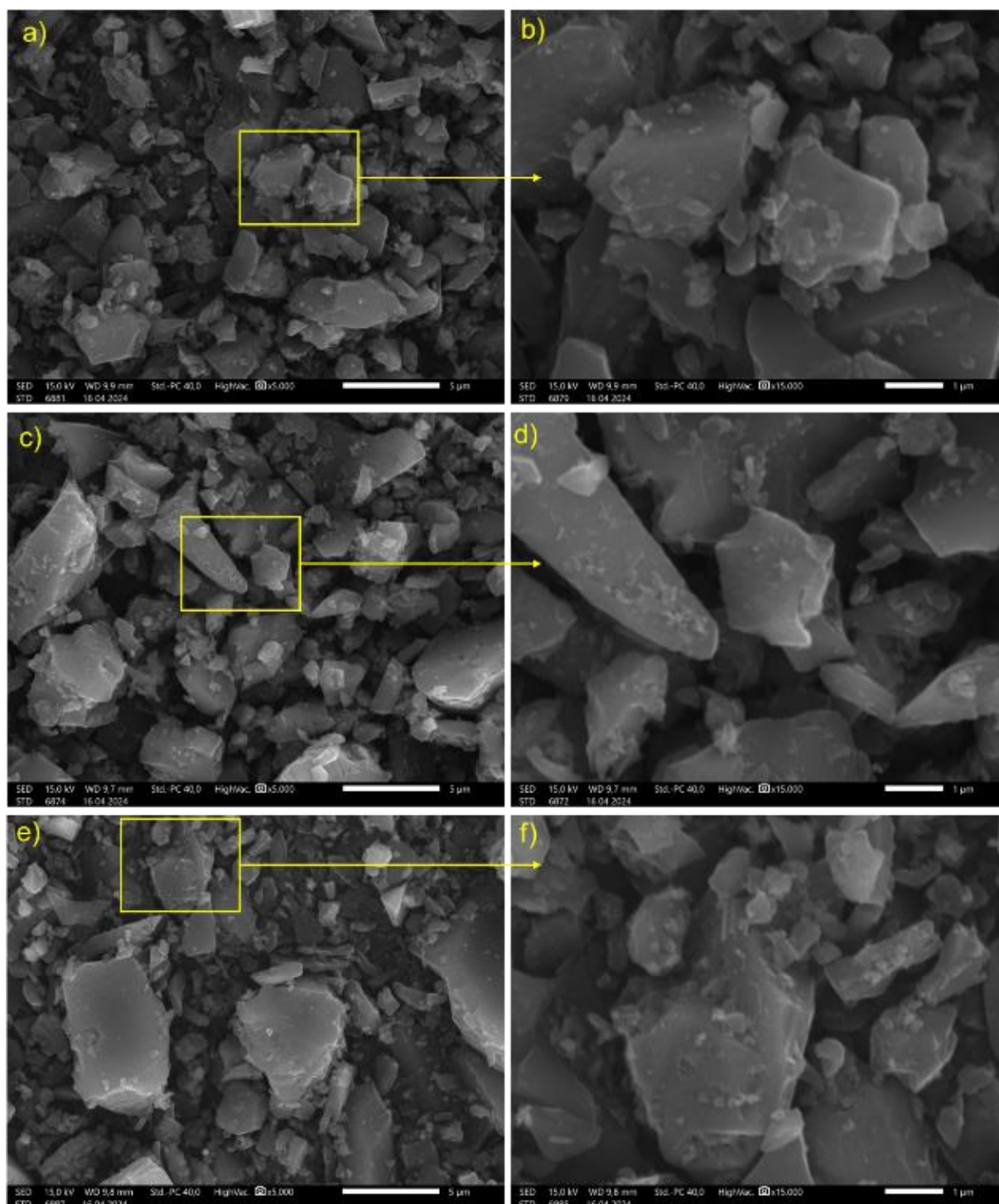
**Figure 2.** (a)  $N_2$  gas absorption profile of activated carbon material, and (b) pore diameter vs cumulative volume of MPL peel samples.

The specific surface area of carbon electrodes MPL-300, MPL-500, and MPL-700 can be calculated by the BET method with values of  $411.311 \text{ m}^2/\text{g}$ ,  $378.786 \text{ m}^2/\text{g}$ , and  $266.822 \text{ m}^2/\text{g}$  with total pores of MPL-300, MPL-500, and MPL-700 of  $0.149 \text{ cm}^2/\text{g}$ ,  $0.136 \text{ cm}^2/\text{g}$ , and  $0.090 \text{ cm}^2/\text{g}$  respectively. These results indicate that MPL-300 has the highest specific surface area of  $411.311 \text{ m}^2/\text{g}$ . However, high surface area is not always a determining factor in increasing the specific capacitance value of carbon electrodes. This is because the higher surface area of MPL-300 can reduce the active properties of carbon which will cause brittle properties on the electrode, which can inhibit electrochemical properties [17].

The surface morphology of banana peel from all concentration variations is illustrated in Figure 3. Figures 3 (a) and 3 (b) show the morphology of MPL-300 samples at 5000x and 15000x magnifications. The MPL-300 sample

displays a surface dominated by particle chunks and carbon aggregates in the size range of 291 – 612 nm at 5000x magnification and 130 – 353 nm at 15000x magnification. Figures 3 (c) and (d) show the morphology of MPL-500 samples at 5000x magnification with a size range of 361 – 1519 nm and at 15000x magnification the size range is 213 – 770 nm. The addition of a chemical activator of 500 mmol/g  $ZnCl_2$  resulted in a relatively larger aggregate surface size. Figures 3 (e) and (f) with the MPL-700 sample show the morphology at 5000x magnification with a size range of 188 – 800 nm and 116 – 465 nm at 15000x magnification.

The elemental content for the three electrodes was evaluated through EDS. The results of the EDS characterization for the MPL-300, MPL-500, and MPL-700 samples were in the form of percentages of the constituent elements of the banana peel carbon electrode shown in Table 2.



**Figure 3.** SEM images carbon of peels (a) MPL-300 magnification 5000x, (b) MPL-300 magnification 15,000x, (c) MPL-500 magnification 5,000x, (d) MPL-500 magnification 15,000x, (e) MPL-700 magnification 5,000x, (f) MPL-700 magnification 15,000x.

**Table 2.** Percentage content of elements in MPL peel samples.

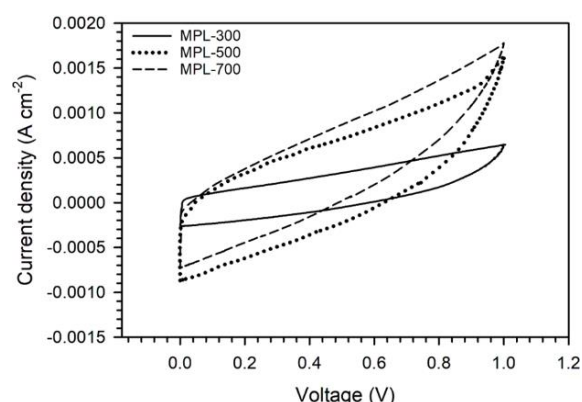
Sample code	Percentage of elements (%)			
	C	O	P	B
MPL-300	90.32	9.59	0.09	-
MPL-500	88.74	10.38	0.22	0.66
MPL-700	86.79	13.06	0.15	-

Based on Table 2, it can be seen that the composition of the elements of the banana peel carbon electrode is carbon (C), oxygen (O), phosphorus (P), boron (B), and nitrogen (N). The elements of the banana peel content are dominated by carbon content with a percentage of 86.79% – 90.32%. The percentage of oxygen



content in MPL-300, MPL-500, and MPL-700 are 0.59%, 10.38%, and 13.06% respectively. The percentage of phosphorus content in the MPL-300, MPL-500, and MPL-700 samples is 0.09%, 0.22%, 0.15%, followed by the boron content in MPL-500 which is 0.66%, in the MPL-300, MPL-500, and MPL-700 samples there is no nitrogen content.

In this case, oxygen is one of the heteroatoms and self-doping elements which is a contribution from the basic elements of biomass waste [18]. The high O element significantly presents an increase in the wettability and hydrophilicity features of the material which dramatically encourages electrolyte infiltration which causes faradaic redox reactions in the electrode material [19]. Phosphorus, boron, and nitrogen are natural dopings that can produce pseudocapacitor properties which means it is possible to achieve higher specific capacitance and energy density in supercapacitor electrochemical testing [20].

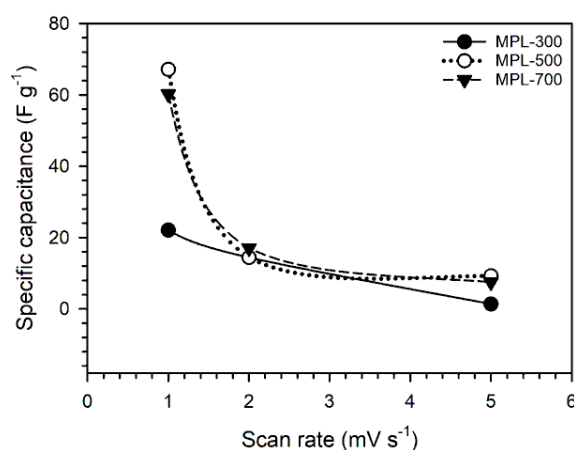


**Figure 4.** CV curve of MPL peel sample.

The electrochemical properties of activated carbon from supercapacitor electrodes were measured through CV shown in Figure 4. The results of the CV measurement are in the form of a curve with a distorted rectangular shape indicating that the supercapacitor has normal EDLC properties [21]. The area of the curve formed between the charge current ( $I_c$ ) and the discharge current ( $I_d$ ) indicates the specific capacitance value produced by the carbon electrode. The larger the area of the curve formed, the greater the specific capacitance value produced [22]. CV testing was carried out

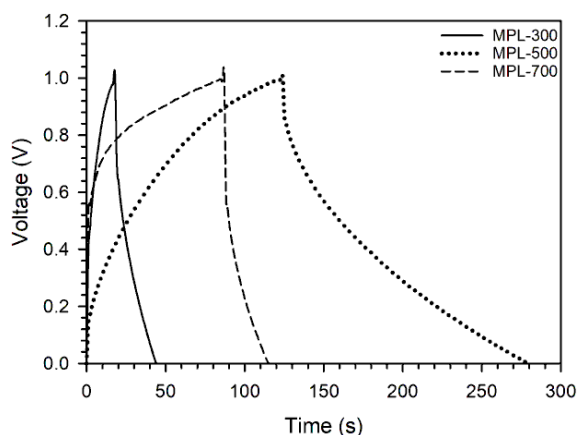
with a constant scan rate of 1 mV/s. CV testing on MPL peel carbon electrodes using  $H_2SO_4$  electrolyte solution as a source of ion charge.

The MPL-300 and MPL-700 samples have the smallest CV curve area compared to the MPL-500 sample which has the largest curve area. These results indicate that the MPL-500 sample has the highest specific capacitance value and the largest curve so it has the ability to store the largest electrochemical energy among other samples. The concentration of MPL-300 can produce a specific capacitance value of 22.06 F/g. The addition of the concentration of  $ZnCl_2$  activator to 500 mmol/g can significantly increase the specific capacitance value to 67.14 F/g with an energy density of 9.325 Wh/kg and a power density of 33.60 W/kg. The addition of the activator concentration reaching 700 mmol/g can reduce the specific capacitance value to 60.21 F/g because the addition of higher concentrations can reduce the physical properties of the electrode such as carbon content. This shows that 500 mmol/g has the optimum specific capacitance value.



**Figure 5.** Specific capacitance curve versus scan rate.

Furthermore, the specific capacitance at higher scan rates is shown in Figure 5. From the figure, it can be seen that the specific capacitance decreases with the high scan rate given. This is because at high scan rates, the electrolyte ions do not have enough time to enter the pores of the electrode [23].



**Figure 6.** GCD curve of MPL peel sample.

More detailed electrochemical properties analysis evaluated through GCD can be seen in Figure 6. All samples show isosceles triangle curves indicating normal double-layer electrochemical properties [24]. The MPL-500 sample shows the longest charge-discharge

measurement time which is marked by a larger isosceles triangle curve shape compared to the MPL-300 and MPL-700 samples. This proves that the MPL-500 sample has very good electrochemical properties resulting in a relatively higher specific capacitance value compared to the MPL-300 and MPL-700 samples. This is in accordance with the CV measurements which are also confirmed by the SEM results where the MPL-500 displays the sample morphology at 5000x magnification with a size range of 361 – 1519 nm and at 15000x magnification the size range is 213 – 770 nm. The administration of a chemical activator of 500 mmol/g  $\text{ZnCl}_2$  produces a relatively larger aggregate surface size. In EDS, the addition of 500 mmol/g  $\text{ZnCl}_2$  activator concentration can increase the percentage of carbon elements to 81.65%.

**Table 3.** Comparison of specific capacitance values of several biomasses

Biomass	Activator	Csp (F/g)	References
Ricinus communis shell	KOH	85	[25]
Palm waste	$\text{CaCl}_2$	137	[26]
Kapok	KOH	31.9	[27]
Sechium edule leaves	KOH	114	[28]
Orange peel	KOH	144	[29]
MPL-500	$\text{ZnCl}_2$	67.14	In this study

Table 3 shows a comparison of several specific capacitance values of several biomass with the addition of several chemical activators. Activated carbon made from MPL peels chemically activated using  $\text{ZnCl}_2$  has the potential to be used as a supercapacitor cell electrode.

## CONCLUSION

Multidoped carbon O, N, P, and B based on *Musa paradisiaca L.* peel waste has been successfully prepared using variations in  $\text{ZnCl}_2$  activator concentrations of 300 mmol/g, 500 mmol/g and 700 mmol/g. The activated carbon obtained has a high carbon content and a good amorphous structure. MPL-500 activated carbon has optimal physical properties with a carbon content of 81.65%, oxygen 17.39%, phosphorus 0.42%, and boron 0.52%.

Electrochemical performance was evaluated using 1 M  $\text{H}_2\text{SO}_4$  electrolyte with a two-electrode system. The highest specific capacitance produced reached 67 F/g with an energy density of 9.325 Wh/kg and a power density of 33.60 W/kg. Furthermore, this study shows the use of MPL peel waste as a porous carbon-based material to be used as a supercapacitor electrode. Therefore, this research can be a reference for obtaining biomass-based porous carbon with environmentally friendly synthesis for supercapacitor energy storage devices.

## ACKNOWLEDGMENTS

This research is financially supported by the first-year project of the Fostered Village Program (PDB) at the Institute for Research and Community Service, the University of Riau

with the title "Solid biomass waste processing based on smoke-free and environmentally friendly technology for carbon production in Sumber Sari Village, Tapung Hulu District, Kampar Regency" with contract number of 1007/UN19.5.1.3/AL.04/2024.

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