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# Conversion of *Salam* leaves (*Syzygium polyanthum* (Wight) Walp.) bio-kitchen waste as functional activated carbon for sustainable supercapacitor electrodes

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**Abstract.** Porous carbon based on bio/organic waste is a very popular raw material used in high-level applications due to its advantages of high porosity, high electrical conductivity, suitable pore structure, and good stability. In this study, bio-kitchen waste is used as a precursor to obtaining high functional activated carbon which is applied to electrochemical energy storage applications. This kitchen waste is focused on Salam leaves (*Syzygium polyanthum* (Wight) Walp.). Precursors are converted to porous carbon through a simple technique and a green approach without the addition of synthetic materials. In addition, activated carbon is designed in a new form of monolith without a binder. The material properties were thoroughly investigated through monolith dimension reduction and X-ray diffraction. The dimensions of the monolith are reviewed based on mass, thickness, and diameter. The activated carbon obtained shows porosity and amorphous properties which are useful in supporting its electrochemical natures. Furthermore, the electrochemical properties of carbon electrodes were reviewed using standard cyclic voltammetry and galvanostatic charge-discharge methods in a two-electrode system. In addition, a 1M H<sub>2</sub>SO<sub>4</sub> aqueous electrolyte was selected to enhance the supercapacitor cell performance and it exhibit high specific capacitance of 145 F g<sup>-1</sup>. Based on these results, it is surprising that bio-kitchen waste has great potential as a high-carbon material for high-level applications.

## 1. Introduction

One of the serious environmental and economic pollution problems facing the world today is kitchen waste. Kitchen waste is waste with very complex components such as a mixture of food scraps, bones, meat, water, a mixture of leftover kitchen spices, plastic, tissue, oil, and salt. In general, bio-kitchen waste dominates 24% to 49% of household waste. Several western countries especially America and Europe produce almost about 120 kg per capita, which is almost 30% of the total food production of these countries [1]. This is also experienced by countries in Asia including China and Indonesia as countries with very high population numbers. In addition, their proportion is expected to continue to grow along with population growth and social development. Furthermore, the composition of kitchen waste is more complicated than that of livestock and agricultural waste where kitchen waste is relatively crumbly, smells, contains components of parasites, pathogenic microorganisms, and other

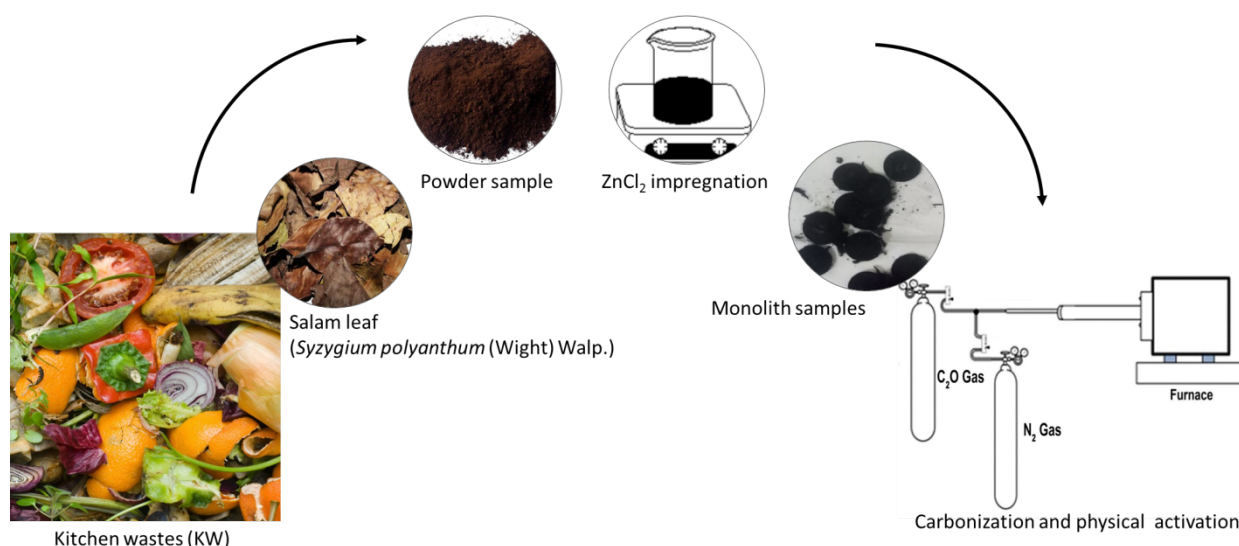


harmful substances thus it is very important to control and overcome them with appropriate methods, techniques, and approaches and efficient. Several countermeasures for kitchen waste have been performed and reported such as the use of their organic materials for the production of animal feed and organic fertilizers. In addition, their rich potential in sugar and protein allows further developments such as the production of biodiesel, methane, hydrogen, and ethanol as fuel sources [2]. On the other hand, kitchen waste, especially organic waste, has a high lignocellulose content, which has the potential as a high carbon source, especially kitchen spice waste. Several previous reports have shown the potential of bio-kitchen waste as a high carbon source for various applications such as waste curcumin as a based material for li-ion batteries [3], ginger as porous carbon for supercapacitor energy storage [4], and lemongrass for reinforcing applications [5]. In the last decade, porous carbon is in great demand by researchers as a based material for electrodes for energy storage applications, especially supercapacitors. This is due to the outstanding properties of carbon including high electrical conductivity, low internal resistance, good chemical stability, and the availability of abundant and renewable raw materials. On the other hand, the utilization of bio-kitchen waste has been applied to supercapacitor applications such as ginger [6], bilimbi [1], curcumin [7], carrot waste [8,9], vegetables [10], etc. They demonstrated the potential of bio-kitchen waste as high carbon with varied structures including nanofibers, nanosheets, hierarchical pore structures which are very much needed to improve the high performance of supercapacitors. However, in the synthesis of carbon materials, they still use multi-step techniques, the addition of metal oxides or conduction polymers, and the use of binders. This can significantly reduce renewability, have mass production costs and produce risky by-products.

In this research, bio-kitchen waste is converted into activated carbon through a new approach, without conduction polymer and metal oxide for supercapacitor application. Furthermore, bio-kitchen waste is focused on the organic waste of *Salam* leaves (*Syzygium polyanthum* (Wight) Walp.). Activated carbon was synthesized from *Salam* leaves by using chemical impregnation of zinc chloride at a concentration of  $0.5 \text{ mL}^{-1}$  at high-temperature pyrolysis. The pyrolysis process is performed in an  $\text{N}_2$  and  $\text{CO}_2$  gas environment consisting of carbonization and physical activation. Material properties confirm high amorphous structure and good porosity. Furthermore, their electrochemical properties show a relatively high specific capacitance of  $145 \text{ F g}^{-1}$  in the  $1 \text{ M H}_2\text{SO}_4$  aqueous electrolyte. Finally, these results provide important information in the management of bio-kitchen waste especially *Salam* leaves (*Syzygium polyanthum* (Wight) Walp.) at high levels as a carbon source for supercapacitor energy storage applications.

## 2. Materials and Methods

*Salam* leaves were chosen as biomass precursors collected from restaurants in Pekanbaru City, Riau Province. Next, the *Salam* leaves are cleaned, dried, and cut into small pieces. The dried precursor was pre-carbonized at  $250 \text{ }^\circ\text{C}$  for 2.5 hours in the oven. Further, the pre-carbonized sample was crushed and ground to obtain a powder sample. To uniform the powder size, the dry precursor was sieved on a  $60 \text{ }\mu\text{m}$  sieve. The dry powder precursors were chemically impregnated by mixing them with a zinc chloride solution in a concentration of  $0.5 \text{ mL}^{-1}$ . The zinc chloride chemical was obtained from Merck KGaA, Index-No: 030-003-00-2, Darmstadt Germany.  $30 \text{ g}$  of precursor dry powder was mixed with  $150 \text{ mL}$  of zinc chloride solution which was stirred using a hotplate at  $80 \text{ }^\circ\text{C}$  at  $300 \text{ rpm}$ . The precursor was then dried in a vacuum oven at  $110 \text{ }^\circ\text{C}$ . Furthermore, the precursor powder sample was converted into a monolith without the addition of a binder through a hydraulic press.  $0.7 \text{ g}$  of powder was put into a  $2 \text{ cm}$  diameter mold pressed using a hydraulic press with a mass equivalent to  $8 \text{ tons}$ . A total of  $15$  monolith samples were pyrolyzed at high temperatures consisting of carbonization and physical activation. Carbonization begins at a temperature of  $30 \text{ }^\circ\text{C}$  to  $600 \text{ }^\circ\text{C}$  in an  $\text{N}_2$  gas environment, then physical activation from a temperature of  $600 \text{ }^\circ\text{C}$  to  $800 \text{ }^\circ\text{C}$  in a  $\text{CO}_2$  gas environment. For comparison, samples were also prepared under conditions of  $\text{N}_2$  carbonization only. Finally, the sample was neutralized using distilled water. To facilitate data analysis, the samples were labeled SL800 (sample pyrolyzed completely via carbonization and physical activation) and SL-precursor (sample pyrolyzed via carbonization only).



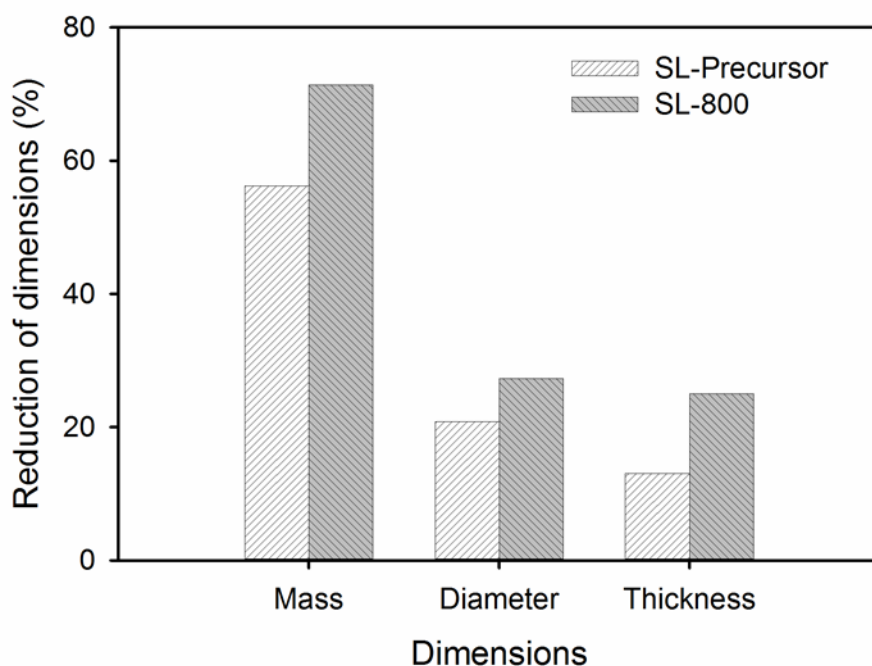
**Figure 1.** Preparation of porous carbon derived *Salam* leaf as functional activated carbon for sustainable supercapacitor electrodes

Activated carbon samples were completely characterized through material properties and electrochemical properties. The material properties of activated carbon were evaluated by reducing monolith dimensions including mass, thickness, and diameter. Furthermore, these shrinkage dimensions are used to calculate the monolith density. Monolith density is calculated based on the standard equation [11]. The electrochemical properties were reviewed through a two-electrode system in an aqueous electrolyte of 1 M H<sub>2</sub>SO<sub>4</sub>. 97% H<sub>2</sub>SO<sub>4</sub> chemical obtained from Panreac Quimica S.A.U. The supercapacitor cell is arranged in a stack of two solid without added the binder material which bounded by a separator. The sample of solid coins is based on activated carbon from *Salam* leaves while the separator is based on organic membranes from duck eggs [12]. In general, electrochemical properties were evaluated through cyclic voltammetry and galvanostatic charge-discharge techniques. The cyclic voltammetry technique was evaluated in the 0-1 V voltage range at different scan rates covering 1 to 10 mV s<sup>-1</sup>. Specific capacitance is determined based on the standard equation [13]. Furthermore, the galvanostatic charge-discharge technique was evaluated at a constant current density of 1.0 A g<sup>-1</sup>. Specific capacitance, energy density, and power density are evaluated completely through standard formulas [14,15].

### 3. Results and discussions

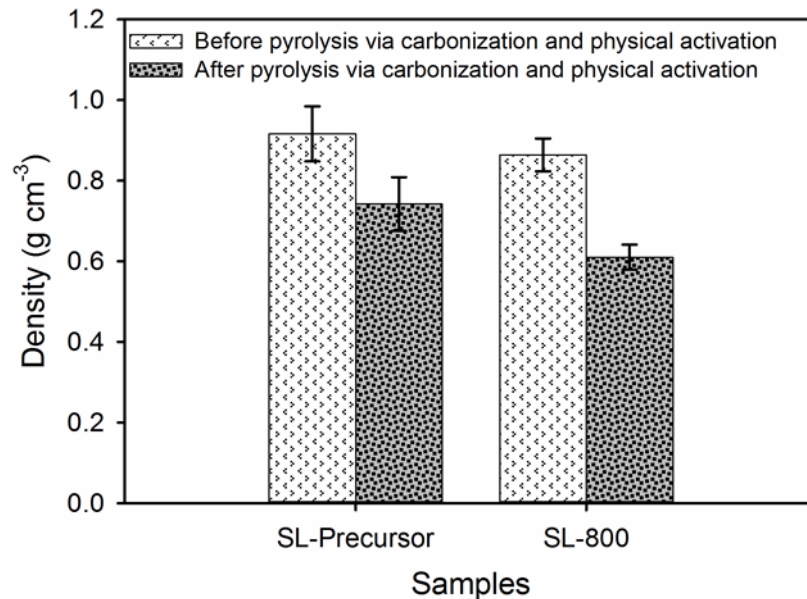
The binder-free monolithic forms of activated carbon were evaluated in detail for their dimensional reduction in the pyrolysis process via carbonization and physical activation [16]. The dimensions of the monolith including mass, thickness, and diameter changed values after high-temperature pyrolysis. This is because the pyrolysis process evaporates non-carbon compounds and degrades lignocellulosic compounds [8,17]. The reduction in mass, thickness, and diameter of the monolith sample without binder are shown in Figure 2. The mass dimensions show the highest reduction of 56.25-71.42% followed by diameter and thickness of 20.81-27.27% and 13.04-25.00%, respectively. This confirms that the pyrolysis process has successfully evaporated the impurity compounds and produced carbon fixed. Furthermore, the SL-800 sample showed a higher dimensional reduction than the SL-Precursor sample, indicating that the high-temperature activation evaporated more non-carbon compounds. The SL-Precursor which was pyrolyzed at 600 °C in an N<sub>2</sub> gas environment was considered to have not maximized the evaporation of non-carbon compounds and it had not initiated good pore formation. In addition, the chemical impregnation of ZnCl<sub>2</sub> was not significantly impregnated in the sample. Moreover, the chemical impregnation of ZnCl<sub>2</sub> reacts completely with carbon at high temperatures, resulting in the formation of more oxides and evaporation in the form of H<sub>2</sub>O and CO<sub>2</sub> in the SL-800

sample. The combination of chemical activation and physical activation in the SL-800 sample significantly maximizes the development of high porosity in the monolith sample. It resulted in the reduction of the dimensions of the monoliths increasing drastically especially their average mass.



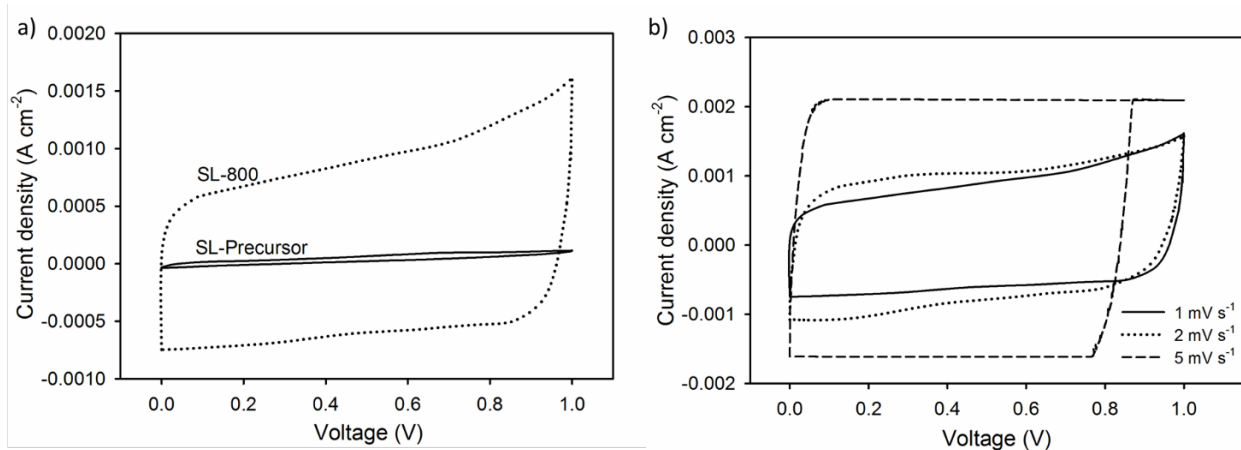
**Figure 2.** Reduction of dimensions for SL-Precursor and SL-800

Furthermore, the density of the monolith was also evaluated intensely in the high-temperature pyrolysis process, as shown in Figure 3. In general, the density of the SL-Precursor and SL-800 monolith samples degraded after the pyrolysis process via carbonization and physical activation from  $0.9163 \text{ g cm}^{-3}$  and  $0.8641 \text{ g cm}^{-3}$  to  $0.7424$  and  $0.6105 \text{ g cm}^{-3}$  with a mean standard deviation of  $\pm 0.061$ . The density value obtained in this study is almost the same as several other studies that have been previously reported, especially the material from coconut husk [18], pineapple leaves [19], and banana stem [20]. The decrease in the density value initiates relatively better pore formation through carbonization and high-temperature physical activation [21]. However, the SL-Precursor samples only showed a density degradation of 18.5% indicating their relatively poor pore formation. Furthermore, the carbonization process that produces tar and solid ash as by-products inhibit the optimization of the pore development of the monolith sample [22]. This is very unfavorable to the performance of the supercapacitor electrodes. On the other hand, the SL-800 sample significantly showed a larger relative density degradation of 25.0% indicating a better porosity development than the SL-Precursor sample. In addition, complete pyrolysis via carbonization and high-temperature physical activation has maximally reduced non-carbon compounds and removed solid tar and ash in monolith samples. This property is very advantageous in improving the electrochemical properties and high performance of supercapacitor electrodes, especially based on porous carbon-based materials from waste biomass. This result is confirmed to be relatively similar to the previous study [13]. In detail, this was confirmed in the evaluation of the electrochemical properties by means of cyclic voltammetry and galvanostatic charge-discharge which are discussed next.



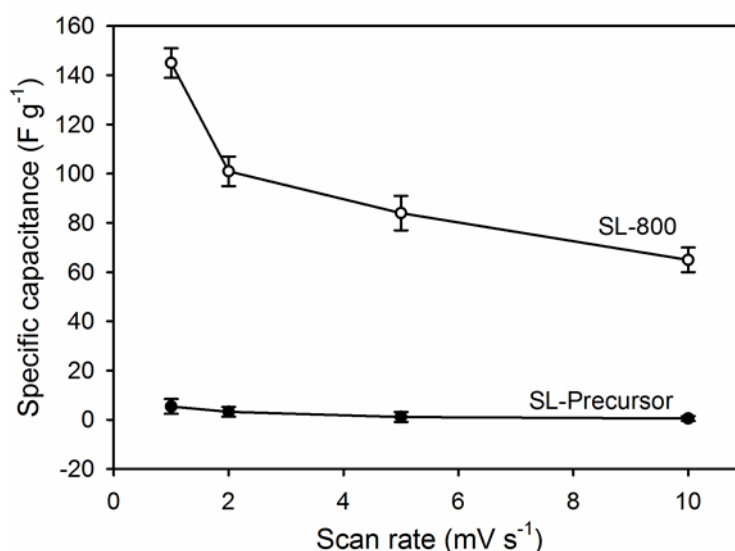
**Figure 3.** The density of SL-Precursor and SL-800 samples

The electrochemical properties of the monolith functional activated carbon derived from *Salam* leaves were evaluated in a two-electrode system. The supercapacitor cell is composed of two solid electrodes in the form of a circular surface bounded by a separator. The separator is selected through the latest approach from eggshell membrane which is considered to have stable permeability properties and is environmentally friendly. Furthermore, an aqueous electrolyte was applied to confirm the electrochemical properties in a  $1 \text{ mL}^{-1}$  sulfuric acid ( $\text{H}_2\text{SO}_4$ ) solution. Moreover, the capacitive properties were evaluated in detail through cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) techniques. Figure 4a shows the CV curves of the SL-Precursor and SL-800 samples. In general, SLs samples show a quasi-rectangular curve which indicates the electrochemical double layer (EDLC) properties [23]. In addition, in Figure 4(a) there is no apparent pseudocapacitance effect confirming the sample has high porous carbon. This is considered normal especially porous carbon from organic waste [24,25].



**Figure 4.** (a) CV curve of SL-Precursor and SL-800 samples, (b) CV curve in different scan rate ( 1, 2, and  $5 \text{ mV s}^{-1}$ ) of SL-800 samples

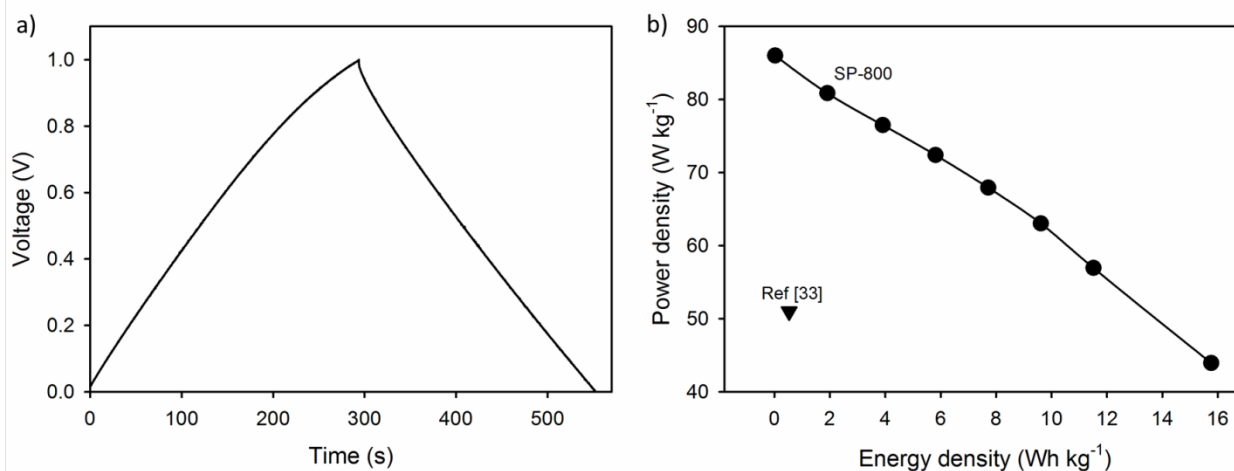
The closed-loop of the CV curve confirms the capacitive nature of the sample. The sample SL-800 drastically shows a large hysteresis loop indicating high capacitive properties whereas SL-Precursor shows an almost flat hysteresis loop confirming a relatively weak capacitive property. Based on the standard equation, the specific capacitance for the SL-Precursor and SL-800 samples was  $5.4 \text{ F g}^{-1}$  and  $145.5 \text{ F g}^{-1}$ , respectively. The combination of chemical activation of  $\text{ZnCl}_2$  and physical activation of  $\text{CO}_2$  at high-temperature pyrolysis has succeeded in increasing the capacitive properties of porous carbon-based supercapacitors from *Salam* leaves waste, as shown in sample SL-800. In contrast to the SL-800 sample, the SL-Precursor sample only has a capacitive property of  $5.4 \text{ F g}^{-1}$ . This is because their process which is only pyrolyzed in the  $\text{N}_2$  carbonization cannot produce material properties that support the performance of the electrode, especially on the porosity properties. This has been thoroughly confirmed on monolith dimensional reduction and density degradation previously. Furthermore, the SL-800 sample has high porosity properties so that it can increase the specific capacitance from  $5.4 \text{ F g}^{-1}$  to  $145.5 \text{ F g}^{-1}$ . However, the capacitive properties in this study are relatively the same as several other studies of different biomass precursors such as mangosteen [26], coffee grounds [27], bamboo, *Terminalia catappa* leaves [28], and bamboo [11]. Figure 4(b) shows the CV curve of the sample SL-800 at different scan rates from  $1 \text{ mV s}^{-1}$  to  $5 \text{ mV s}^{-1}$ . The CV curve still shows a quasi-rectangular shape which indicates that the SL-800 sample has good EDLC properties. These results are relatively the same as some previous studies [11,29]. However, higher scan rates can significantly reduce the capacitive properties of the SLs samples, as shown in Figure 5. Scan rates greater than  $1 \text{ mV s}^{-1}$  to  $10 \text{ mV s}^{-1}$  reduce the SL-Precursor sample-specific capacitance of  $5.4 \text{ F g}^{-1}$  to  $0.8 \text{ F g}^{-1}$  as well as SL-800 samples from  $145.5$  to  $65.3 \text{ F g}^{-1}$ . This case is considered normal for a porous carbon based material [30].



**Figure 5.** Specific capacitance vs. scan rate curve of SL-Precursor and SL-800 samples

The deeper electrochemical behavior of the SL-800 sample was evaluated by using galvanostatic charge discharge (GCD). The GCD profile for the SL-800 sample at a constant current density of  $1.0 \text{ A g}^{-1}$  is shown in Figure 6(a). It clearly shows a symmetrical triangular shape confirming the satisfactory electrochemical reversibility. Furthermore, the distinctive triangular shape further confirms that the electrode exhibits the characteristics of a double layer capacitor and excellent electrochemical reversibility [31,32]. Based on the standard equation, the specific capacitance obtained is  $166 \text{ F g}^{-1}$ . In addition, a relatively small ohmic drop ( $iR$  drop) of  $14 \text{ m}\Omega$  was observed, indicating high electro-conductivity and low internal resistance. Moreover, energy density and power density are also evaluated in a fully Ragone plot. Figure 6(b) shows the Ragone plot for the SL-800 sample. The maximum energy density was obtained at  $15.7648 \text{ Wh kg}^{-1}$  in  $1 \text{ M H}_2\text{SO}_4$  electrolyte and

it resulted in a power density of  $86.02 \text{ W kg}^{-1}$ . This values were relatively higher than the previously reported with precursor biomass of European deciduous trees with energy density of  $0.53 \text{ Wh kg}^{-1}$  at power density of  $51 \text{ W kg}^{-1}$  [33].



**Figure 6** (a) GCD profile of SL-800, (b) Ragone plot of SL-800

#### 4. Conclusion

High porous carbon based on bio/organic waste was successfully obtained from kitchen waste of *Salam* leaves (*Syzygium polyanthum* (Wight) Walp.) as electrode material for supercapacitor. Bio-kitchen waste of salam leaves was converted into activated carbon by using multi-activation of chemical activation of  $\text{ZnCl}_2$  and physical activation of  $\text{CO}_2$  gas atmosphere. Zinc chloride was prepared in a  $0.5 \text{ mL}^{-1}$  solution. Furthermore, the effect of high-temperature pyrolysis of  $\text{N}_2$  carbonization and physical activation of  $\text{CO}_2$  was studied in more depth. The precursor sample which was only pyrolyzed in  $\text{N}_2$  carbonization showed relatively lower porosity compared to the SL-800 sample which was completely pyrolyzed ( $\text{N}_2$  carbonization and  $\text{CO}_2$  physical activation). This significantly impacts the electrochemical properties of the supercapacitor. In the two-electrode system, the SL-800 sample displayed significantly higher capacitive properties than the SL-Precursor sample of  $166 \text{ F g}^{-1}$  at a current density of  $1.0 \text{ A g}^{-1}$ . Furthermore, the resulting energy density is relatively high at  $15.7648 \text{ Wh kg}^{-1}$  in  $1 \text{ M H}_2\text{SO}_4$  electrolyte. Finally, these results confirm the high potential of bay leaf bio-waste as a porous carbon source for the high application of supercapacitor energy storage devices.

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