Power generation from palm kernel shell biochar in a direct carbon fuel cell

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Received: 6 December 2019 / Accepted: 5 February 2020 / Published online: 12 February 2020
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Abstract

The diminishing fossil fuel reserves and energy shortage are serious challenges that have directed the search for alternative fuel sources for energy generation in the twenty-first century. Biomass-based carbon materials are alternative and renewable fuel sources used for contemporary energy conversion and present a promising solution to the above crisis. This study investigates the possibility of power generation from oil palm kernel shell (PKS) biochar through direct carbon fuel cell (DCFC) technology. The objective of the study is to investigate the potential of PKS biochar as a fuel source and consolidate information on the association between the structure of PKS biochar and its electrochemical performance in a DCFC. The effect of acid pre-treatment on the structure of biochar is also investigated. The samples are characterized on the basis of chemical composition (proximate/ultimate analysis) and DCFC performance in terms of open circuit voltage, power densities and electrochemical impedance spectroscopy. The results indicated that carbon, ash and oxygen functional groups on the carbon surface have a major impact on the fuel performance in the DCFC. At DCFC operating temperature of 850 °C, the pre-treated PKS biochar exhibits a maximum power density of 3.3 mW cm⁻² which exceeds that of untreated PKS biochar and activated carbon. Besides high carbon and reduced ash in the biochar, the results suggest that the existence of oxygen functional groups in the pre-treated PKS biochar also plays a crucial role in cell performance by providing additional active sites for electrochemical reaction.

Keywords Renewable fuel source · Energy conversion · Power generation · Palm kernel shell · Biochar · Direct carbon fuel cell

1 Introduction

The conventional sources for energy generation come from non-renewables such as coal, natural gas and petroleum with coal accounting for almost forty-per cent of global power generation. However, the current coal-firing processes are inefficient (25–35% efficiency) and contribute a major bulk towards greenhouse gas emissions [1]. Consequently, efforts are underway to find alternative fuel sources for energy generation which are less detrimental to the ecology and environment. The search for alternative

energy sources has led the way to the utilization and application of biomass-based sources. Biomass is an alternative energy source which accounts for 14% towards the global annual energy consumption. It is also the fourth major energy scheme in line after the conventional energy sources [2]. Biochar fuel production from abundantly available biomass opens avenues to utilize it as a renewable energy source for power generation along with tackling the issue of waste utilization. The research thus aims to build off-grid power generation with low impact on the

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environment utilizing biomass from oil palm waste as a potentially sustainable future energy source.

Fuel cells are an efficient technology which transform chemical energy to electrical energy through electrochemical oxidation reaction. Majority of the fuel cells are based on the utilization of gaseous fuels such as hydrogen or carbon monoxide which do not occur naturally for power generation. These fuels are typically produced through processes such as coal gasification or natural gas reformation. Competing with these conventional fuels are the solid carbon fuels, which occur naturally and are renewable. In a direct carbon fuel cell (DCFC) technology, the solid carbon's stored chemical energy is directly converted into electrical energy via electrochemical oxidation reaction. The DCFC efficiency could reach to 80% (cumulative product of theoretical, voltage and fuel utilization efficiencies) and the overall system efficiencies have been expected to be over 60–70% [1]. The DCFC can be grouped into three categories basis electrolyte type which mainly includes molten hydroxide, molten carbonate and solid ceramic electrolyte YSZ (yttria-stabilised zirconia). Molten carbonate and molten hydroxide electrolytes suffer from serious problems such as electrolyte stability, degradation and leakage as the electrolytes are corrosive [3–5]. DCFC systems operating on solid oxide electrolytes have been the focus of research as they offer fuel flexibility and the set-up is simple and uncomplicated [6]. The DCFC set-up with solid oxide electrolyte, operating on solid carbon fuel also presents a non-corrosive environment compared to the other two electrolytes. Higher efficiencies of energy conversion from carbon fuels could be achieved through the DCFC. Subsequently, ways to improve the electrochemical performance of fuels at the anode is one of the major hindrances of their application in the DCFC. Therefore, various surface treatment approaches such as acid, alkali, heat, laser, and plasma treatments, have been usually employed on the carbon materials to enhance their electrochemical performance [7–10].

To our information, there has been no research work done on the association between physicochemical properties of oil palm PKS biomass through surface modification and their electrochemical output in a DCFC. Thus, this study carries out a relative analysis of the performance of pre-treated PKS carbon fuels with untreated PKS carbon fuel and commercial activated carbon (AC) fuel in the DCFC system. In this study, pre-treatment of PKS biomass using hydrochloric acid (HCl) was employed for surface modification. Subsequently, the correlation between the characteristics of carbon fuels and electrochemical output was investigated. The significance of this research lies in defining the potential of waste PKS biomass as an

alternative fuel source in the DCFC. The importance of the research also lies in making DCFC technology a viable, cleaner alternative to traditional power generation methods.

2 Materials and methods

Palm kernel shell (PKS) biomass was collected from an oil palm plant in Sri Ulu Langat in Dengkil, Selangor, Malaysia and was utilized for biochar preparation. The PKS biomass was treated with HCl and the biochars were designated as 'PKS-HCl', while the untreated PKS biochar is referred to as 'U-PKS.' Commercial activated carbon (AC) was bought from Sigma-Aldrich (M) Sdn Bhd Malaysia along with other chemicals.

2.1 Fuel preparation

A suitable pyrolysis temperature to produce maximum biochar yield from PKS was first determined. Pyrolysis temperature in the range of 400–600 °C was investigated. The PKS biomass samples underwent pyrolysis in a horizontal split quartz tube (HST 12/400, Carbolite) reactor at a heating rate of 10 °C min⁻¹ in N₂ atmosphere (99.9%) flowing at 1 L min⁻¹. Pre-treated PKS was produced using HCl concentrations ranging from 0.1 to 4.0 M [7] after which it was dried in the oven (UN75, Memmert, USA) according to the ASTM D2867-09 standard method (drying at 105 °C for 24 h). The biochar production from pre-treated PKS was carried out under the optimum pyrolysis temperature by following the same pyrolysis method as listed above.

2.2 Fuel characterization

The weight loss in biochars was examined on thermogravimetric analyser (TGA) (STA 6000, Perkin Elmer thermo-balance, USA). 10 mg of the sample was heated from 30 to 900 °C at a heating rate of 10 °C min⁻¹ under N₂ as inert gas. At the temperature of 900 °C, inert gas supply was cut off and switched to oxygen gas at a flow rate and heating rate of 20 mL min⁻¹ and 10 °C min⁻¹, respectively. The samples were held at this stage for 20 min for ash determination [11]. CHNS analysis was conducted to deduce the biomass and biochar elemental compositions. The CHNS analysis was conducted by using CHNS analyser (Elementar Vario MACRO Cube, Germany) to measure carbon, nitrogen, hydrogen and sulphur values. Higher heating value (HHV) was determined from element compositions with the help of Eq. (1) [12]:

$$\text{HHV}(\text{MJ/kg}) = 0.3491\text{C} + 1.1783\text{H} + 0.1005\text{S} - 0.1034\text{O} - 0.0151\text{N} - 0.0211\text{A} \quad (1)$$

where C, N, H, O, S and A represents carbon, nitrogen, hydrogen, oxygen, sulphur and ash contents of materials, respectively [12]. The values are expressed in weight per cent on a dry basis.

2.3 DCFC design and electrochemical performance analysis

The DCFC performance test of HCl treated PKS biochars, U-PKS and AC at different operating temperatures was conducted by placing 0.1 g of the biochar sample on the button cell. The button cell was placed between the anode and cathode ceramic tubes. DCFC contains a cathode (LSM), anode (Ni-YSZ) and electrolyte (YSZ) procured from Ningbo SOFCMAN Energy Technology Co. Ltd., China. Silver wire with high conductivity was used for current collection. The DCFC operating temperature was controlled by temperature controllers and electric heaters. The anode side was supplied with N₂ gas (flow rate of 600 mL min⁻¹) and the cathode side was supplied O₂ gas (flow rate of 200 mL min⁻¹). The operating temperature of DCFC was examined at 750 °C, 800 °C and 850 °C. The voltage output (referred to as open-circuit voltage, OCV) was measured. All electrochemical assessments were done using a Potentiostat (Gamry, Interface 1000E, Germany). The electrochemical tests were started after the OCV reached a steady state. The current–voltage (*I*–*V*) curve of the run was evaluated through electrochemical terminal with a scanning degree of 10 mV s⁻¹ and an excitation voltage amplitude of 30 mV. The electrochemical impedance spectroscopy (EIS) was recorded using the Nyquist plot and the cell resistances were calculated at the given temperature in 100 kHz to 0.1 Hz frequency range. The data was recorded for all the biochar samples.

3 Results and discussion

3.1 Biochar yield

The biochar yields from PKS biomass after pyrolysis are tabulated in Table 1. The yield of produced biochars was examined under different pyrolysis temperatures. As reported, the pyrolysis process conditions namely temperature, gas pressure, purge gas and heating rate significantly influence the biochar yields [13].

It can be seen in Table 1 that the biochar yield is affected by pyrolysis temperature, which is in agreement with other studies [14–16]. As seen in Table 1, the biochar yield in the range of 20–34% is observed for PKS biomass.

Table 1 PKS biochar yield at different pyrolysis temperatures

Biochars	Biomass weight (g)	Biochar weight (g)	Yield (%)
PKS_400	1.00	0.20 ± 0.01	20
PKS_450	1.00	0.32 ± 0.03	32
PKS_500	1.00	0.34 ± 0.02	33
PKS_550	1.00	0.33 ± 0.02	34
PKS_600	1.00	0.28 ± 0.01	28

Increasing pyrolysis temperature from 400 to 550 °C is seen to increase the yield of PKS biomass, giving the highest yield of 34% at 550 °C. Based on these results as shown in Table 1, pyrolysis temperature of 550 °C is selected to produce PKS biochar, owing to the highest biochar yield. It has also been reported by researchers that the properties of biochar, such as biochar yield, carbon content, and surface area, are adequately enhanced at around 500 °C when the thermal decomposition of cellulose and hemicellulose is complete [13, 17, 18]. The physicochemical characteristics of biochar such as elemental composition, fixed carbon, ash, volatile matter, surface area and thermal stabilities are dependent on pyrolysis temperatures [19–21].

The biochar yield declines as the pyrolysis temperatures increase. This decrease in char yield is probably caused by the primary decomposition of the biomass at higher temperatures or due to char residues undergoing secondary decomposition [22, 23]. When the biomass is heated at high temperatures, the polymer experiences breakage of chemical bonds, which in turn causes volatile compounds to release. This also leads to reshuffling of reactions which occurs with the residue matrix. Such reactions refer to as primary reactions. Thereafter, these primary reactions give rise to unstable volatile compounds which further convert through secondary reactions. Rise in gas yields and consequent decline in char yield happens mainly when pyrolysis vapours undergo secondary cracking at higher temperatures [24, 25].

Apart from the temperature being an important parameter in the pyrolysis process, the type of feedstock also influences the physicochemical properties of the biochar. In general, woody biomass in most cases has greater contents of hemicellulose, cellulose and lignin compared to non-woody biomass [26]. The higher amount of lignin present in the plant biomass is reportedly responsible for carbonization and hence leads to an increase in the biochar formation [27]. The lignin content of PKS biomass is reported to be approximately 50%, while for palm mesocarp fibre (PMF), empty fruit bunch (EFB), oil palm frond and oil palm trunk is approximately 27%, 22%, 21% and 25%, respectively [28, 29]. The high lignin content in PKS biomass contributes to a high char yield and can be

explained on the fact that lignin contains more benzene rings which forms the char [25]. Hence for the production of solid fuel which could be a potential replacement for coal for power generation, biomass with high lignin is suggested. This also serves as the reason behind selecting PKS biomass as the carbon fuel for DCFC application.

3.2 Proximate analysis

Proximate analysis of raw PKS biomass, PKS biochars and AC was determined using thermogravimetric analysis (TGA) i.e. volatile matter, moisture, fixed carbon and ash contents were estimated. As seen in Table 2, the proportions of fixed carbon (FC) generally increased while volatile matter (VM), moisture, and ash decreased for pre-treated PKS biochars compared to the raw biomass. The biochars were high in carbon content (68.8–72.0 wt%), indicating the pre-treatment step made the fuels more carbonaceous. This property makes the biochars a better reactive fuel in the DCFC system as carbon is the main substrate for electrochemical reactions in the DCFC [30, 31]. Higher content of fixed carbon also corresponds to more chemical energy, i.e. a high calorific value, thus leading to a better DCFC performance [32]. It is also observed from the proximate analysis results that chemical pre-treatment reduced the fraction of VM significantly for the pre-treated biochars which ranged from 18.3 to 23.3 wt% for PKS biochars. Typically, high content of VM in the biochar would reduce its heating efficiency when used as a fuel in the fuel cell [33]. An important fuel property is the fuel ratio which denotes the ratio of fixed carbon against volatile matter. It is a specific value demonstrating the property of carbon fuels and classifying the rank of coals according to ASTM 388.

As seen in Table 2, the results for pre-treated biochars show an increment in the fixed carbon contents and a decline in the volatile matter contents. The increase in

the fuel ratios indicates higher efficiencies during DCFC operation. The PKS biochars had moisture in the range of 3.1–5.8 wt%. The presence of moisture in the biomass is in the form of a mineralized aqueous solution which contains various cations, anions or non-charged species [33]. Alternatively, if the moisture content becomes significantly high after the pre-treatment process, it causes negative effects such as lowering of calorific value, reducing the fuel efficiency and delayed release of volatiles [34, 35]. The ash content which is a crucial parameter in determining the efficiency of the pre-treatment process, is seen to decrease in the pre-treated biochars. Compared to the raw biomass samples, ash was significantly reduced in PKS biochars (7.1–1.0 wt%). A high quantity of ash will become a hindrance for the carbon particles to be in contact with the anode. This will eventually lead to the blockage of active sites of the anode during the electrochemical oxidation reaction, adversely impacting the performance of fuel cell [36]. In specific as shown in Table 2, for PKS biochars, the lowest ash value of 1.0 wt% was seen for biochar which was treated with 2 M HCl (referred here as PKS-2.0). The important outcome of this analysis was that HCl pre-treatment seem to have been effective in reducing the ash content from the raw PKS biomass substantially. Hence for further characterization, PKS-2.0 biochar was chosen.

There have been studies done focusing on pre-treatment techniques by mostly employing different types of coal as carbon fuel. In a study on the demineralization of anthracite coal with dilute HCl, results showed good performance in the DCFC which was attributed to having a high fixed carbon content, medium values of volatile matter and low values of moisture and ash (particularly that of sulphur) [32]. The observations in these studies are consistent with the present study and suggest that the pre-treatment process could assist in obtaining biochars with the desired proximate characteristics.

3.3 CHNS analysis

The elemental composition of PKS biochars assists in evaluating its possible utilization as a fuel in a DCFC system. Comprising a high percentage of carbon suggests that the biochar can be a possible fuel source as carbon is the main substrate for the electrochemical reaction in the DCFC system [37]. The CHNS analysis results of PKS biochars along with AC fuel are outlined in Table 3.

Results presented in Table 3 illustrate that PKS biochars have high a carbon content which indicate that PKS biochars could be potential fuel in the DCFC system. Table 3 also gives the ratio of oxygen to carbon (O/C) ratio for the PKS-2.0 biochar which is 23% (oxygen content is calculated by difference). This suggests the existence of increased oxygen-containing groups present in the biochar [30, 37].

Table 2 Proximate analysis of PKS biochars

Carbon fuels	FC (wt%)	VM (wt%)	Moisture (wt%)	Ash (wt%)
Raw PKS	20.2	67.3	5.4	7.1
AC	92.0	–	5.3	2.7
U-PKS	73.1	24.1	1.7	1.1
PKS-0.1	68.8	23.3	4.8	3.3
PKS-0.5	69.9	22.8	4.5	3.1
PKS-1.0	71.1	20.4	4.1	1.5
PKS-1.5	70.5	21.5	3.7	1.1
PKS-2.0	70.2	21.6	3.1	1.0
PKS-2.5	71.6	19.4	5.0	1.8
PKS-3.0	71.7	19.0	5.5	1.9
PKS-4.0	72.0	18.3	5.8	1.9

Table 3 Ultimate analysis of biochar fuels (wt%)

Proximate analysis	C	H	N	S	O	H/C	O/C	HHV (MJ/kg)
AC	96.4	0.6	0.0	0.6	0.2	0.01	0.01	34.4
U-PKS	75.8	3.1	4.1	2.1	14.2	0.04	0.18	27.3
PKS-2.0	76.4	3.2	2.1	0.2	17.9	0.04	0.23	28.9

The electrochemical oxidation of biochar is reported to be enhanced by the high concentration of oxygen-containing groups. The surface oxygen functional groups refer to the presence of the reactive sites which enhances the rate of electrochemical oxidation reaction for carbon fuels [30]. These ratios refer to oxygen functional groups and are much higher when compared to untreated biomass and to commercial carbons such as activated carbon (8.8%), carbon black (3.5%) and graphite (1.1%) [37]. Table 3 also shows that PKS-2.0 biochar contains low fractions of sulphur (S) which is a component present in ash. The low content of contaminants especially sulphur is very favourable for effective working of the DCFC [38]. It is essential to examine the effect of ash components on the DCFC performance as the gathered ashes would impact the working and lifespan of DCFC [38–40].

There are limited studies describing the influence of carbon fuel impurities on the working of DCFC. Li et al. performed acid pre-treatment on coal and showed the removal of the mineral impurities from coal which in turn helped in protecting the electrodes and the electrolytes [41, 42]. Weaver et al. studied the effect of ash addition in the DCFC system. They supplemented the electrolytes of a fixed bed DCFC with 10 wt% of fly ash and reported that this addition did not have a significant effect on the polarization behaviour [43]. Vutetakis et al. employed a fluidized bed DCFC to demonstrate the influence of several mineral impurities. They noticed a decline in the current output at high over-potentials. The phenomenon as described by them was caused by the passivation of electrodes which resulted from the formation of an ash film on the surface of the electrode [44]. Cherepy et al. used petroleum cokes in the DCFC which contained sulphur (2.5–6 wt%). They described the corrosion of anode to be responsible for the decline in the cell performance [38]. The higher heating values (HHV) of the PKS-2.0 biochar was determined through elemental values and outlined in Table 3. The HHV values depicted in Table 3 show a relation with the ash content (Table 2). With the decrease in ash content, the HHV increase.

3.4 OCV measurement of pre-treated PKS biochars

Figure 1 gives the open-circuit voltage (OCV) of the tests carried out on AC, U-PKS and PKS-2.0 biochar in the DCFC

Fig. 1 OCV curve for AC, U-PKS and PKS-2.0 biochars**Table 4** OCV values for AC, U-PKS and PKS-2.0 biochars

Biochars	OCV (V)	Power density (mW cm ⁻²)
PKS-2.0	0.81 ± 0.02	3.50
AC	0.81 ± 0.01	0.71
U-PKS	0.79 ± 0.01	0.50

operating at 800 °C. Table 4 shows the voltages (OCV) obtained from PKS-2.0 biochar, U-PKS biochar and AC. PKS-2.0 biochar gave the similar voltage of 0.81 V with AC. U-PKS biochar gave a slightly lower voltage than AC (0.79 V) as depicted in Table 4. During OCV testing, it was observed that there was a sharp rise in the OCV values at a DCFC operating temperature of 500 °C, which probably occurred as a result of the ionic conduction movement in the phase of the electrolyte [4]. As the DCFC operating temperature increased to 800 °C, it was seen that the OCV values became stable. This is due to the increase in the rate of ionic conduction of the electrolyte and the electrochemical reactions at the two electrodes [38, 42, 44, 45]. Looking at the CHNS analysis results of the PKS-2.0 biochars in Table 3, it is seen that the PKS-2.0 biochar contains a high amount of elemental carbon, recorded at 76.4 wt%. This might contribute to better performance in DCFC.

3.5 Current and power densities of HCl treated PKS biochars

The selected PKS-2.0 biochar sample was studied to evaluate its performance at 750 °C, 800 °C and 850 °C in the DCFC system and the results are presented in Fig. 2. The polarization curves of fuel cell voltage versus the current density shown in Fig. 2a and appear to be alike in their shapes and decrease from the maximum voltage of roughly 0.81 V. The PKS-2.0 biochar sample exhibits a maximum power density of 0.6, 2.9 and 3.3 mW cm⁻² at 750 °C, 800 °C and 850 °C. This higher power density production could be attributed to a higher O/C ratio of 0.23, and a lower ash content of 1.0 wt%. In previous research, Li et al. used AC and carbon black as fuel supplies for DCFC. They treated both these carbon fuels with HNO₃ to improve their surface properties. The results indicated that treated fuels showed higher electrochemical reactivity which was due to the existence of surface oxygen functional groups. It was concluded that these surface oxygen functional groups translate into free reactive sites which enhances the anodic discharge rate in the DCFCs [7]. In another research study, HCl treatment was done on coal samples to be used as fuels for DCFCs. The acid-treated coal samples presented the highest power densities of 115 mW cm⁻² at

800 °C. The study concluded that the main reason behind this elevated performance was the presence of oxygen functional groups which convert into reactive sites for the electrochemical oxidation reaction. It was also concluded that the improved surface area of the acid-treated coal samples could have contributed towards the power density increment but was not the major contributor for the high power density output [41].

3.6 Impedance study in HCl treated PKS biochars

The electrochemical impedance spectroscopy (EIS) measurements were performed on PKS-2.0 biochar at DCFC operating temperatures of 750 °C, 800 °C and 850 °C and the curves are presented as impedance spectra in Fig. 3a–c. These figures help to establish the solution resistance of the electrolyte which is the high-frequency

Fig. 2 **a** I - V curve, **b** I - P curve of PKS-2.0 biochar for DCFC operated on 750 °C, 800 °C and 850 °C

Fig. 3 EIS spectra for DCFC. **a** 750 °C, **b** 800 °C, **c** 850 °C obtained for PKS-2.0 biochar

intercept of the real axis (R_s) and the polarization resistance (R_p) which is the low-frequency intercept of the real axis. In all the Nyquist plots, the resistances decrease with the rise in the DCFC operating temperature. This signifies that the lowering of the resistance leads to a higher power density output. A study which utilized carbon black fuel generated a power density output of 13.0 mW cm^{-2} with a low resistance at 900°C compared to the cell operated at 700°C with a lower power density output at 3.6 mW cm^{-2} and higher resistance [46]. Specifically for PKS-2.0 biochar, the values for solution resistance, R_s , are 7.15, 0.43 and $0.25 \Omega \text{ cm}^2$ at 750°C , 800°C and 850°C , respectively. The polarization resistance, R_p , was obtained at 44.3, 41.2 and $31.4 \Omega \text{ cm}^2$ at 750°C , 800°C and 850°C , respectively.

4 Conclusion

The research study establishes the potential of waste oil palm-based palm kernel shell (PKS) biochar as a prospective alternative fuel source for power generation in a direct carbon fuel cell (DCFC). PKS biomass was pre-treated with HCl for structural modification. Subsequently, the association between the physicochemical properties after structural modification and electrochemical performance of the PKS as fuel source for DCFCs was examined. The major changes in the structure and physicochemical properties of the acid treated PKS fuels included high carbon content, reduced ash content and additional surface oxygen functional groups which influences the electrochemical performance of PKS fuels in the DCFC. The maximum power density of 3.3 mW cm^{-2} was obtained at a DCFC operating temperature of 850°C , giving the least total cell resistance. The outcome, therefore, advocate that the structure of the carbon fuel plays an essential role in DCFC performance and structural modification of the biochars might be a practical approach to improve the applicability of the fuels in DCFC systems.

Acknowledgements The authors would like to thank for the financial support given by Taylor's University research Grant Scheme (TRGS/ERFS/2/2016/SOE/003) and University Kebangsaan Malaysia Grant Scheme DIP-2018-012 and GUP-2018-013.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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