

## PHYSICOCHEMICAL PROPERTIES OF HYDROCHARS PRODUCED FROM *KHAYA SENEGALENSIS* LEAVES USING HYDROTHERMAL CARBONISATION

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### Abstract

Dried leaves of *Khaya senegalensis* were converted into hydrochars via hydrothermal carbonisation (HTC) at different temperatures (200, 225, and 250 °C) under three residence times (24, 48, and 72 h). This study was conducted to investigate the effect of temperature and residence time on the production of hydrochars. The physicochemical properties of hydrochars produced from *Khaya senegalensis* dried leaves using HTC were also analysed. Experimental results showed that when HTC temperature was increased, a significant decrease in hydrochar yield was observed, along with increased energy content and yield. The highest energy content of 28.813 MJ/kg was obtained when the dried leaves were heated at 250 °C for 72 h, which was 62.72% of increased energy content compared to that of raw dried leaves. The pH values of the hydrochars ranged between 5.0 and 6.8, which were slightly acidic. The scanning electron microscopy results showed that hydrochars have higher porosity and surface area compared to raw dried leaves. The highest surface area of 13.92 m<sup>2</sup>/g was obtained at 250 °C, with 24 h of residence time. It was found that the physicochemical properties of hydrochars varied significantly with variations in temperature and residence time during hydrothermal carbonisation.

Keywords: Hydrochar, Hydrothermal carbonisation, *Khaya senegalensis* leaves, Physicochemical properties.

## 1. Introduction

The conversion of biomass waste (dead plants or animals) into beneficial carbon material (coal, bio-oil) through hydrothermal carbonisation (HTC) has gained global attention due to its ability to produce hydrochars. The conversion of biomass waste into hydrochars has helped in minimising the amount of waste released into the environment as this method can successfully turn waste into wealth. The promising properties of hydrochars have promoted their utilisation for various purposes, such as activated carbon application, catalyst synthesis, and CO<sub>2</sub> adsorption [1-4].

Hydrochars can also be used as soil fertilisers, as well as in carbon sequestration to decrease the climate change phenomena. HTC is an attractive thermochemical method of converting biomass into hydrochars. During HTC, temperature and reaction time play big roles towards the properties of hydrochars [2, 5-7]. It was found that the biomass would undergo huge physicochemical and molecular alterations, which include mass loss, structural transformation, and adjustment in surface charges (pH). Various studies have been conducted that employed low cost and abundant sources, such as tree leaves [1, 6], Chinese fan palm [8], acacia [7], grass [9], and eucalyptus [10].

Chen et al. [11] and Kang et al. [12] described the properties of hydrochars produced from watermelon peels and corn stalk, respectively. Xu et al. [13] reported the propitious conversion of phoenix tree leaves into carbon-enriched hydrochars using the microwave-assisted HTC. Jaruwat et al. [14] studied the effects of HTC temperature and reaction time on cattail leaves. They proposed that these parameters can greatly affect the surface area and morphology of the resulting hydrochars. Saqib et al. [6] produced hydrochars via HTC using dried leaves, which attained the highest energy content of 19.98 MJ/kg after being heated at 240 °C. They employed short residence time which was 30 minutes, and the reaction was carried out using stainless steel with a nozzle at top for gas collection. However, the sort of dried leaves used was not specified, leaving possibility for more research into their potential as hydrochar via hydrothermal carbonization.

Thus, this study was conducted in the interest of finding out the potential of *Khaya senegalensis* dried leaves as hydrochars synthesised using the cost-effective method of hydrothermal carbonisation. *Khaya senegalensis* is commonly used as medicinal treatment for various illnesses in Africa, but this species has never been studied as a potential source of hydrochars. The leaves of this species contain mostly mannose sugar, which is derived from glucose and has different chemical compounds, namely, alkaloids, flavonoids, sterols, triterpines, saponins, glycosides, and carbohydrates [15].

*Khaya senegalensis*, from the Meliaceae family, also known as African Mahogany, are planted alongside car parks and roads in the Universiti Sains Malaysia, Engineering Campus. These trees were originally planted to provide a greener environment, as well as to provide shade. The fallen leaves, however, were subjected to normal disposal. Thus, turning the dried leaves into useful hydrochars will lead to a maximum waste-to-wealth conversion. Therefore, this study was conducted to turn the fallen dried leaves of *Khaya senegalensis* into hydrochars by configuring their physiochemical properties. These hydrochars were produced under different temperatures and residence times to optimise these properties. However, the hydrochars produced in this study are not available for specific applications as further work is still in progress.

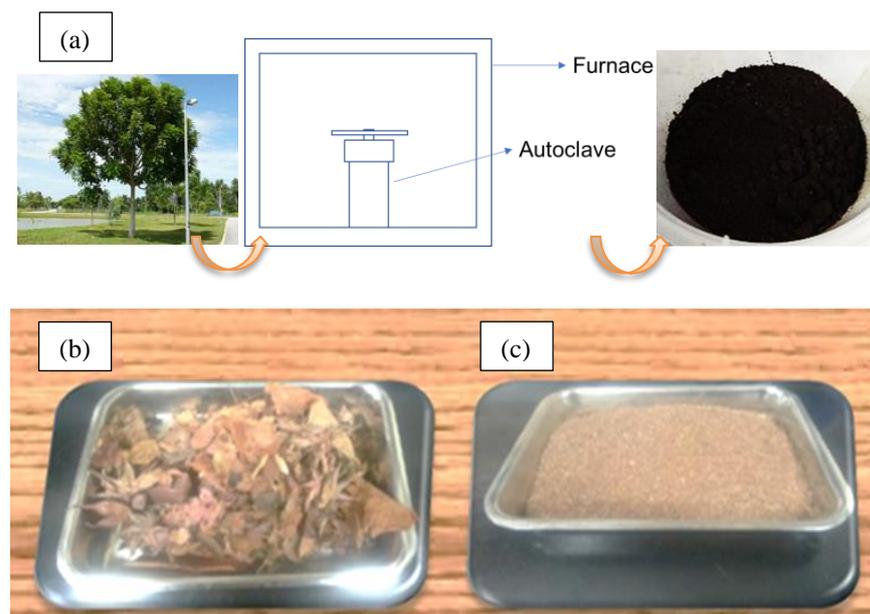
## 2. Material Preparation

Dried leaves of *Khaya senegalensis*, found abundantly around the Engineering Campus of USM, were gathered. The leaves were then crushed into smaller sizes and passed through sieves to yield particles with a size of lesser than 2 mm. Figure 1 shows images of the dried leaves before and after being crushed, respectively. Then, the crushed leaves were parched in an oven at 110 °C prior to the carbonisation step. Roughly 10 g of feedstock was dissolved in 35 ml of hot deionised water at a ratio of 1:7 (w/v).

This procedure was performed using the laboratory-scale stainless steel autoclave [1]. Due to simpler configuration of reactor with no nozzle at top for gas collection, the residence time required is far longer as compared to the previous work done by Saqib et al. [6]. The samples were physically mixed to confirm proper wetting. These samples were then heated at three different temperatures (200, 225, and 250 °C) at different residence times (24, 48, and 72 h).

Approximately 50% of feedstocks turned into hydrochars at approximately 200 to 250 °C. These samples were labelled as HC-residence time-temperature, for example, HC-24-200 denoted the hydrochar sample synthesised for 24 h of residence time at 200 °C.

After each HTC process, the autoclave was taken out from the oven and placed in water for quenching reaction at 25 °C. The resulting hydrochar was washed with acetone and placed in an oven at 105 °C for 2 h for subsequent drying [6]. Acetone was used as the cleaning agent in this study because it is water miscible and will not interact with the hydrochar.



**Fig. 1. Flow diagram of turning *Khaya senegalensis* leaves into hydrochar: (a) via HTC; (b) uncrushed dried leaves; and (c) crushed dried leaves.**

## Physical and Chemical Characterisation

The pH of the samples was determined by immersing 1 g of hydrochars into 20 ml of distilled water. The solution was stirred for 1.5 h before sampling was done for pH measurement using the CyberScan pH 1500 from Eutech Instruments. The final hydrochar yield was calculated from the amount of biomass, as shown by the following Eq. (1):

$$\text{Yield} = \frac{\text{mass of hydrochar}}{\text{mass of biomass}} \times 100\% \quad (1)$$

An elemental analyser (Perkin Elmer 2400, Series II) was used to determine the existence of C, H, N, S, and O elements in the hydrochars. The Brunauer-Emmet-Teller (BET) surface area analysis of all hydrochar samples was conducted using the Micromeritics ASAP 2020. The surface morphology of the dried leaves, before and after undergoing HTC, was analysed using the Scanning Electron Microscope (SEM) Quanta FEG 650, with acceleration voltage of 15-20 kV. The energy yield of the hydrochar samples was measured using an advanced bomb calorimeter, CAL2K. The calculations for energy densification ratio and energy yield were based on Eq. (2) and Eq. (3), respectively [6]:

$$\text{Energy densification ratio} = \frac{\text{Energy content of hydrochar}}{\text{Energy content of feedstock}} \quad (2)$$

$$\text{Energy yield} = \text{Yield of hydrochar} \times \text{energy densification ratio} \quad (3)$$

## 3. Results and Discussion

### 3.1. pH measurement

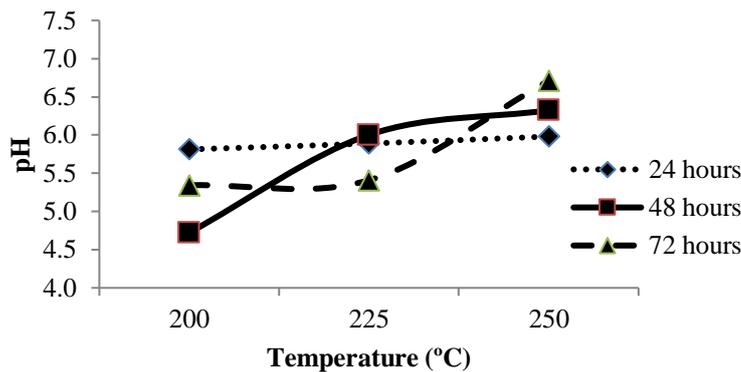
#### 3.1.1. Effect of HTC temperature on pH

The raw dried leaves of *Khaya senegalensis* had a pH of 4.8, which was acidic. Meanwhile, Fig. 2 shows that the pH values of the hydrochar samples are 5.8, 5.8, and 5.9 at 200, 225, and 250 °C, respectively, after undergoing the HTC process for 24 h. These results showed that the increased HTC temperature has led to the pH values being increased by 20.25%, 21.82%, and 23.72% for HC-24-200, HC-24-225, and HC-24-250 samples, respectively. These results are in line with previous studies that reported increases in HTC temperatures that led to increased pH values [5, 16]. *Khaya senegalensis* leaves contain mostly mannose sugar, which is derived from glucose, and this glucose undergoes a hydration process, leading to the formation of sugar monomers. Consequently, this formation of monomers increases the development of H<sup>+</sup> ions on the surface, which will eventually raise the pH from 4.8 to 5.9. Nizamuddin et al. [17] suggested that an increase in pH indicates the creation of slightly acidic functional groups on the hydrochar surface. Another possibility for this observation is that the arrangement of OH<sup>-</sup> ions in the liquid phase during HTC occurs much faster than the establishment of H<sup>+</sup> ions, leading to higher pH values at higher temperatures.

#### 3.1.2. Effect of residence time on pH

Another parameter that could affect the hydrochar properties is residence time. Thus, residence time ranging from 24 to 72 h was implemented, with 24 h intervals. After undergoing 48 h of HTC process, the pH values of the samples had increased from 5.0 to 6.0 and 6.3 at 200, 225, and 250 °C, respectively. Similar increases

were observed with HC-48-200, HC-48-225, and HC-48-250, whereby the pH values of these samples were increased by 3.7%, 24.17%, and 30.82%, respectively, compared to the raw sample. When HTC was performed for 72 h, the pH values of the samples were 5.3, 5.4, and  $6.7 \pm 0.1$  at 200, 225, and 250 °C, individually. These were increments in pH values at 10.53%, 11.89%, and 38.8% for HC-72-200, HC-72-225, and HC-72-250 samples, respectively. Based on Fig. 2, the pH values of the samples are increased with an increase in temperature for all three residence times, which are 24, 48, and 72 h. Each hydrochar sample possessed a higher pH value than the raw sample. The pH values of the hydrochar samples were ranged between 5.0 and 6.8, which are in agreement with results found in literature. Most hydrothermal products were slightly acidic due to the presence of  $H^+$  ions on the surface of hydrochars [5, 16].



**Fig. 2. pH trends of hydrochars produced at different temperatures and residence times.**

### 3.2. Ultimate analysis

The ultimate analysis revealed the constitution and percentage of elements present in the hydrochars under different temperatures and residence times. The elemental composition was significantly affected by the HTC process. The first primary reaction in the HTC of biomass, namely, solid to solid conversion, would involve condensation, decarboxylation, and drying out reaction, which is when the oxygen, carbon, and hydrogen substances will be extensively diminished. Next, the polymerisation of dissolved intermediates found in the liquid to form solids would occur and the last reaction is carbonisation via intermediate solid coke [5]. Since the dried leaves of *Khaya senegalensis* comprises of glycosides, the removal of carboxyl group and the release of carbon dioxide ( $CO_2$ ) would occur through decarboxylation reaction, while the removal of water would occur via dehydration. During HTC, the natural composition of the biomass will be altered, which precedes hydrochars and can be analysed in terms of the percentage of carbon, hydrogen, oxygen, nitrogen, and sulphur elements. Table 1 shows the elemental analysis results of the raw dried leaves, as well as the hydrochar samples at varied temperatures and residence times, which typically consist of carbon, followed by oxygen, hydrogen, nitrogen, and sulphur.

**Table 1. Ultimate analysis of dried leaves before undergoing hydrothermal carbonisation and the resulting hydrochars at varied temperatures and residence times.**

Temperature (°C)	Residence Time (h)	Samples	C%	O%	H%	N%	S%
-	-	Raw	47.29	44.03	6.57	1.22	0.89
	24	HC-24-200	56.32	35.06	6.56	1.39	0.67
<b>200</b>	48	HC-48-200	59.68	31.65	6.53	1.38	0.76
	72	HC-72-200	60.33	30.87	6.52	1.56	0.72
	24	HC-24-225	63.22	27.75	6.47	1.87	0.69
<b>225</b>	48	HC-48-225	64.74	26.39	6.41	1.79	0.67
	72	HC-72-225	65.78	25.38	6.36	1.77	0.71
	24	HC-24-250	66.71	24.69	6.21	1.69	0.7
<b>250</b>	48	HC-48-250	66.78	24.68	6.12	1.77	0.65
	72	HC-72-250	68.11	23.64	6.03	1.47	0.75

The carbon element in the hydrochars will substantially expand with the increase in residence time at different temperatures. The increment in carbon percentage was due to the drying out and decarboxylation reactions that took place during HTC [5, 8-11]. The carbon content in hydrochars was increased because at temperatures higher than 250 °C, approximately 50% yield was achieved with 60% carbon content for most organic wastes [5]. At each temperature, samples with residence time of 72 h showed the highest carbon content, although the increment with residence time of 48 h was low at approximately 1.09% to 1.99% only. This result indicated that the carbon content was higher over a longer residence time during HTC due to the intensive reactions, which was in line with the previous observation made by Chen et al. [11]. The HC-72-250 sample had the highest carbon content among the other samples. This result proved that carbon elements were higher at higher temperature and longer residence time. These results were comparative to the results in literature, where elevated carbon content was reported due to the increase in temperature, which increased the decarboxylation and drying out reactions of the hydrochars [11-14].

Meanwhile, oxygen tends to gradually decrease with all residence times under each temperature setting. The oxygen content was the highest for HC-24-200 and the lowest for HC-72-250. This difference proved that oxygen content would be lower after a longer residence time and higher temperature. The loss in oxygen content was due to combustion and decarboxylation of the hydrochars. Oxygen was released through combustion, dehydration, and decarboxylation during the HTC process [6-9].

Similarly, the hydrogen content of the hydrochars had also decreased with increased residence time. Table 1 shows that the hydrogen content slowly decreases with increasing residence time under different temperatures. The lowest hydrogen content was recorded at 250 °C after 72 h of residence time. This result was similar to the results of oxygen content, whereby the higher temperature and the longer residence time have attributed to lower hydrogen contents. The decreasing percentage of hydrogen content was due to the dehydration and decarboxylation processes, by which its removal was in the forms of H<sub>2</sub>O and CO<sub>2</sub> [6-9].

The nitrogen content was recorded slightly differently from the carbon and hydrogen contents. Nitrogen content had increased with increasing residence time at 200 and 225 °C. At 250 °C, the nitrogen content started to increase after 24 and 46 h, but gradually decreased after 72 h. The nitrogen content showed non-linear trends, which suggested the consequences of the rise in nitrogen partitioning into the aqueous phase when reaction conditions became harsher, in particular, in an array, as observed by Yao et al. [8]. Although the nitrogen content of hydrochars was decreased after 72 h, it was still higher than in the raw dried leaves.

The sulphur content of the hydrochars depicts a decreasing pattern with increases in temperature and residence time, as displayed in Table 1. Each of the three temperature settings shared a similar trend. The percentages of sulphur in the hydrochars and the raw feedstock were very low, which were less than 1%. No significant effect was observed in sulphur content as it was already very low for the raw feedstock, which is parallel to the previous finding by Saqib et al. [6]. The sulphur and nitrogen percentages were lower in hydrochars because they have been dissolved in the preparation fluid during HTC. Results from the elemental analysis were similar to earlier results reported by other studies [1-7, 11-15], whereby carbon has the highest composition in hydrochars, followed by oxygen, hydrogen, nitrogen, and sulphur.

### 3.3. Energy yield and energy content

The energy yield and content of all the synthesised hydrochar samples are listed in Table 2. Energy content describes the total amount of energy accumulated in a material. The energy content of the raw feedstock was 17.125 MJ/kg. At 200 °C and after 24 h, the energy content of the resulting hydrochars was raised to 21.294 MJ/kg. The hydrochar energy content was increased when the temperature was increased, which was parallel to the increase in carbon content. Based on the table, the energy contents of the hydrochar samples are increased when HTC temperature is increased. The energy content of the hydrochars was the highest after undergoing HTC at 250 °C for 72 h, which was 62.72% higher than that of the feedstock. These results were in line with the increase in carbon content.

**Table 2. Energy yield and energy content of the samples.**

Sample	Energy content (MJ/kg)	Mass yield (%)	Energy densification ratio	Energy yield (%)
Raw	17.125	-	-	-
HC-24-200	21.294	62.6	1.243	77.8
HC-24-225	23.724	64.0	1.385	88.7
HC-24-250	26.811	50.4	1.566	78.9
HC-48-200	22.030	61.4	1.286	78.9
HC-48-225	24.541	52.8	1.433	75.6
HC-48-250	25.431	54.0	1.485	80.2
HC-72-200	21.630	63.6	1.263	80.3
HC-72-225	26.525	50.6	1.549	78.4
HC-72-250	28.813	46.0	1.683	77.4

The ratio between the energy content of hydrochars and the feedstock can be defined as energy densification, with a similar increasing pattern as the temperature

and residence time [6, 7]. The energy densification of hydrochar samples is shown in Table 2, which intensifies with the rise in temperature. This trend was equal with the trend of the energy content. The minimum energy densification of 1.243 was recorded at 200 °C after 24 h, while the maximum of 1.683 was recorded at 250 °C after 72 h. These results showed that shorter residence times and lower temperatures would lead to lower energy densification, while a longer residence time and a higher temperature can lead to a higher energy densification. This phenomenon is governed by the increase in temperature and residence time, which will increase the carbon content and consequently, leads to a higher energy content [13].

After 24 h of HTC, the energy yield had increased from 77.8% to 78.9% as rise in temperature from 200 °C to 250 °C, which then decreased to 78.9% at 250 °C. The increase in energy yield from 200 °C to 225 °C is most likely owing to an increase in energy densification, as shown by Eqs. (2) and (3) before. When the temperature is raised to 250 °C, the mass yield declines, and the energy yield falls to 78.9%. Because of the increasing rate of hydrochar formation and unreacted waste, mass yield consistently dropped as temperature rose, with 224°C being the optimum temperature for converting dried *Khaya senegalensis* leaves into hydrochar in this study.

After 48 h, the energy yield began to decrease from 78.9% to 75.7%, which then increased to 80.2%. Similarly, after 72 h, the energy yield had decreased from 80.3% to 78.4%, and decreased again to 77.4%. As a result, the maximum energy yield was achieved at 225 °C after 24 h of HTC, which was due to the highest energy content and energy densification values.

These results were comparable to a previous study on acacia, which showed 87% of energy yield at 200 °C of HTC [7]. As a result, the high-energy hydrochars developed by this research might be used as solid fuels, as well as to make fuel cells, electrode supercapacitors, and as a medium for converting and storing energy, such as batteries. Additionally, hydrochars that are predominantly derived from plant biomass could be considered as a good potential for solid fuel due to their low ash content. These components would include biomass combustion, which can cause harmful outflows, as well as fouling, slagging, and erosion in the combustors. Hydrochars as a fuel source is projected to be eco-friendlier and can be an alternative to coal.

### 3.4. Surface area

Figure 3 shows the specific surface areas of raw dried leaves and hydrochar samples at different temperatures and a constant residence time of 24 h. Residence time of 24 h was chosen because a longer residence time would result in a higher pH for the hydrochars, which would then result in a lower surface area. Pauline et al. [5] reported that surface area would decrease with an increase in pH. The results indicated the increase in surface area, both when using BET and Langmuir methods. Both techniques implied that HTC had successfully altered the surface area of the hydrochars. However, the use of as-produced hydrochars as adsorbents in an industry is slightly ineffective compared to activated carbons. Activated carbons with improved performance characteristics have been proven to be more efficient due to the bigger porosity yield from carbonisation compared to hydrothermal carbonisation [1].

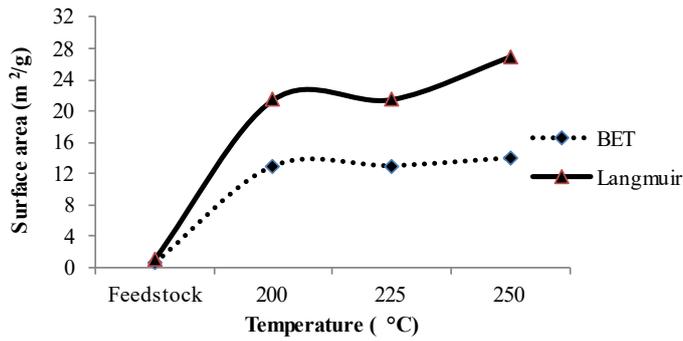


Fig. 3. Surface area analyses of hydrochars using BET and Langmuir methods.

### 3.5. Morphological characteristics

The surface morphology of hydrochars under different HTC conditions was observed using SEM. Figure 4 shows SEM images of raw dried leaves and hydrochars treated for 24 h, with 10,000 times magnification.

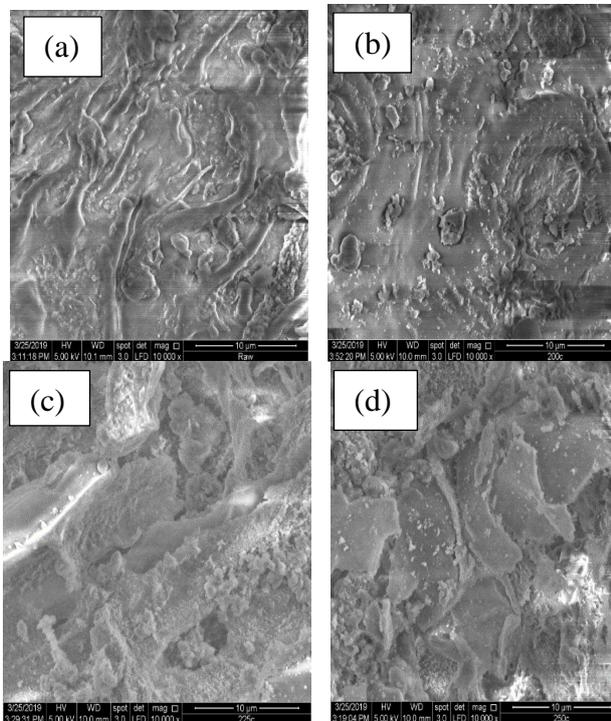


Fig. 4. SEM images: (a) raw feedstock; (b) hydrochar of HC-24-200; (c) HC-24-225; and (d) HC-24-250 at 10,000 times magnification.

From the figure, the surface of the hydrochars appears rougher compared to the surface of the raw dried leaves. This could be because when the leaves underwent HTC process, polymers, such as hemicellulose and cellulose were simply decomposed into portions that led to an irregular and porous surface on the biomass.

These small segments would then melt and form carbonaceous spheres on the surface. Conferring to Jaruwat et al. [14], the surface of hydrochars is likely to appear coarser and rougher at higher temperatures, with unusual microspheres.

These spheres would ease the diffusion of water in the matrix of the *Khaya senegalensis* dried leaves and break the cellulose and hemicellulose bonds, which would indirectly raise the surface area. The untreated dried leaves presented with a uniformed line structure with limited porosity. However, the hydrochar surface appeared to have a huge number of porous structures, and the microstructures of the hydrochars were different than the dried leaves. This observation showed that the degradation of organic components had occurred during the HTC process.

#### 4. Conclusion

Dried leaves of *Khaya senegalensis* were converted into hydrochars via hydrothermal carbonisation (HTC). The biomass to water content ratio was set at 1:7. The results showed that temperature and residence time can affect the properties of the resulting hydrochars. Hydrochars that have pH values ranging from 5.0 to 6.8 can be used as an amendment to reduce the acidity of soil. The synthesised hydrochars possessed comparatively high energy contents compared to the raw material. While the energy content and energy yield were amplified, the mass yield was decreased. The energy content was at its highest at 250 °C and 72 h of hydrothermal carbonisation. The high energy content of hydrochars enables it to become an alternative for fuel, as it produces less harmful gas to the environment and contains less ash. The BET and SEM analyses proved that hydrochars have a higher surface area and porosity compared to dried leaves. The high porosity of hydrochars produced at 250 °C worked well in holding dampness in soils and extended water holding capacity compared to raw feedstock. Hydrochars produced at this temperature also have a high carbon content, which can provide better combustion compared to the raw dried leaves. Therefore, it can be concluded that dried leaves were successfully converted into hydrochars, which were suitable for a wide range of applications, either as soil enrichments or fuel.

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