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Article

# Torrefaction of Lawson Cypress Cone Biomass, Bituminous Coal and Their Blends for Industrial Usage

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#### **Abstract**

Biochar from heat pretreatment of Lawson cypress cone (LCC) biomass, which in this case is a renewable energy source, will enhance its calorific energy value and hydrophobicity to replace coal, a nonrenewable energy source in thermal power plants and metallurgical processes. As such, some associated adverse effects of using coal (viz., bituminous coal, BC), including emissions, costs and availability are to some extent, checked. For that reason, the present study aimed at subjecting 20  $\mu$ m particle size BC, LCC and their equal, 3:1 and 1:3 blends (in form of Samples I-V) to torrefaction in a retort furnace between 0-225 min and 0-300 °C to produce char whose respective mass yields, energy density, fixed carbon content and weight loss would enable their selection for particular industrial application. In terms of optimal torrefaction temperature and residence time in the furnace, Sample I-V in that order, are the best. Due to 13.3 g low weight loss or 13.3% low mass yield of sample V (i.e. 100% LCC), it is suitable for co-firing bioenergy plant. The blends can go into thermal power plants, filtration and metallurgical processes due to their higher energy density and improved combustion characteristics, as obtained in this study. Therefore, LCC is an important biomass that needs to be characterize further for specific utilization to produce biochar of specific grade and industrial usage.

#### **Keywords**

Lawson cypress cone, Torrefaction, Bituminous coal, Biochar, Co-firing, Mass yield

## **Article History**

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#### 1. Introduction

Torrefaction is a thermochemical process that involves heating biomass, coal, or their blends in the absence of oxygen to produce a solid fuel with improved properties for combustion or gasification [1]. Lawson cypress cone (LCC, Chamaecyparis lawsoniana) biomass and bituminous coal (BC) can be subjected to torrefaction individually or in various blends to enhance their energy density, grindability and storage stability [2]. Torrefied biomass and coal can have lower moisture and volatile content, leading to reduced emissions during combustion and can be used in existing power plants without significant [3] modifications to the infrastructure [4,5]. However, challenges that may hinder its acceptance and usage include: cost of torrefaction equipment and the energy required for the process [3], varying quality of torrefied material based on the feedstock composition and torrefaction conditions [6], logistics and handling requirements during its integration with existing coal-based processes and the market acceptance of torrefied biomass and coal as an industrial fuel source [7]. Some unique qualities of BC that may make it advantageous for torrefaction are its higher energy density compared to sub-bituminous and lignite coals, where which torrefaction further increase this energy density, making it a more efficient fuel for combustion; its lower inherent moisture content compared to lower-rank coals, may experience a more substantial reduction in moisture during the torrefaction process [8]; its higher carbon content than sub-bituminous and lignite coals, contributing to higher calorific value and improved combustion characteristics after torrefaction; its lower volatile matter content compared to sub-bituminous coal further reduced by torrefaction, resulting in a more stable and less volatile fuel and; it is more amenable to carbon capture and storage technologies.

In a previous study, Li et al. [9] looked at the co-combustion of specific types of coal and biomass blends as a potential partial or complete substitute for 100% coal utilization in some power plants. Most co-combustion studies assess the performance of different proportions or ratios of the blend based on specific or varied particle sizes, resultant emissions, thermogravimetric analysis, and their heating values during co-firing. For example, Sakuragi & Otaka [10] describe the milling behavior of torrefied biomass and coal blends in a roller mill while Kopczynski & Zuwala [4] explained the energy and mass balances involved in torrefaction, also suggesting that initial refining of biomass by torrefaction before co-combustion is beneficial. While the biomass in question is numerous and diverse, LCC remained to be examined for efficiency and effectiveness in several industrial applications. In view of that, the objectives of this research are to determine the weight loss with temperature (0-300 °C) of crushed LCC biomass, crushed BC, and their mixtures in various ratios in order to produce biochar. Biochar produced can be used for co-firing with coal char, since studies carried out by Lin et al. [11] attest to its production via low-temperature torrefaction. Similar to this present study, Akinrinola et al. [12] researched the torrefaction of four woody biomass at 270-290 °C for 30-60 min to test its suitability for large-scale power stations, where it was discovered that the energy density of the woods improved significantly from 19.2-21.2 MJ/kg. At the same time, Ajikashile et al. [13] examined the properties of torrefied sorghum and millet straw (≤5 mm particle size) at 230, 240, 250, 260, and 270 °C temperature and 30 and 60 min residence time. Apart from the 60 min residence time usually employed [14,15], this study examined the biochar weight produced over an extended time range. Depending on the raw biomass type, torrefaction has already been carried out in fixed-bed reactors, microwaves, fluidized-bed systems, and muffle furnaces [16,17]. This study novelty lies in the investigation of torrefaction effects on various blends of LCC biomass and BC in different proportions (100%, 75:25, 50:50, 25:75), providing insights into their performance for specific industrial applications. The study further evaluates the effects of varying residence times (up to 225 min) and temperatures (up to 300 °C), extending the analysis beyond commonly studied ranges to optimize char properties.

#### 2. Methodology

This work experiment was performed in the General Workshop Laboratory in the Department of Chemical Engineering at BUITEMS Pakistan. BC of various grades and LCC biomass was obtained from Sharegh, Balochistan in the same country. Hammer mill, sieve or screen, sieve shaker, electronic balance and Muffle or Retort furnace were among the equipment employed to achieve the set objectives. Basically, torrefaction process involves drying of sample to eliminate moisture, heating at 200-300 °C in a low-oxygen environment (typically < 10% oxygen), followed by cooling [18]. The technique was carefully carried out in the laboratory.

#### 2.1 Material Grinding and Sieving

A hammer mill with a fixed rotating disc to which six hammer bars are attached was used to grind BC for a 36 min crushing period. After that, the hammer mill was cleaned carefully so that the debris of coal stock in the mill circular casing was not mixed with LCC. Then, LCC were put into the hammer mill hopper to crush for 60 min. It is observed that the material is thrown out centrifugally and crushed by the action of hammer bars and breaker plates fixed around the circular metal casing. Woven wire mesh sieves or screens were used to sieve 300 g of crushed BC using a sieve shaker in order to obtain particles that would be retained in the sieve of 20  $\mu$ m mesh size. Higher sizes ranging from 75-90  $\mu$ m had been used previously for pulverized coal, as experimented in Panahi et al. [19]. After 15 min of sieving, a collecting tray was used to collect the particle size of 20  $\mu$ m of BC, which was 249 g. Polyethylene box was later used to preserve the collected coal sample. In a similar fashion, 400g of LCC was crushed for 30 min and sieved for 55 min, after which 245 g retained in a sieve of mesh size 20  $\mu$ m was collected. Screening, crushing and milling carried out here

was in accordance with the procedure described in Mamvura & Muzenda [15].

## 2.2 Preparing Different Proportions of Samples

An electronic weighing balance with a digital display was used to measure varying ratios of BC and LCC, as shown in Table 1.

Table 1. Sample blends.

S/No.	Item	Ratio	Name	
1.	BC	100%	Sample I	
2.	BC + LCC	75:25 (%)	Sample II	
3.	BC + LCC	50:50 (%)	Sample III	
4.	BC + LCC	25:75 (%)	Sample IV	
5.	LCC	100%	Sample V	

As specified in Table 1, Sample I is 100 wt.% BC, Sample II is 3:1 BC-LCC blend, Sample III is 1:1 BC-LCC blend, Sample IV is 1:3 BC-LCC blend and Sample V is 100 wt.% LCC. Sample II, III and IV blends were mixed thoroughly using a laboratory mixer.

#### 2.3. Char Production

#### 2.3.1 Coal Char from Sample I

About 20 g of BC sample (100%) was weighed using an electronic balance for heat treatment in a muffle furnace in order to produce coal char. First, the temperature of the muffle furnace was set at 0 °C and powered ON to increase the temperature. At a fixed temperature of 20 °C, the BC sample was allowed to stay for 15 min residence time before being taken out and measured. The experiment was repeated at an increasing 20 °C temperature interval up to a temperature of 300 °C at residence times of 15-225 min, leaving intervals of 15 min. Weights of the char was then recorded at the respective furnace temperature and residence time kept. The torrefied materials were then cooled to ambient temperatures under the inert atmosphere before storage or further processing, so that self-ignition and combustion of solid products were avoided [20]. Relationship between reaction time, residence time and torrefaction temperature is vividly explained by Bergman et al. [5].

#### 2.3.2 Char from Other Sample Blends

The same amount was also measured for Samples II, III, IV and V and the process was repeated. However, Samples II-IV contains different specified ratios of BC and LCC.

#### 2.3.3 Mass Yield Calculation

Mass yield,  $y_M$  was computed according to Equation 1 given by Akinrinola et al. [12] and Ajikashile et al. [13].

$$y_M = \frac{M_{Char}}{M_{feed}} \tag{1}$$

Where,  $M_{feed}$  = dry mass of the untreated biomass (g) and  $M_{Char}$  = mass of the torrefied product (g). Note that  $y_M$  can also be expressed in percentage. The entire experimental task is divided into sample preparation and torrefaction process, as illustrated in Figure 1.

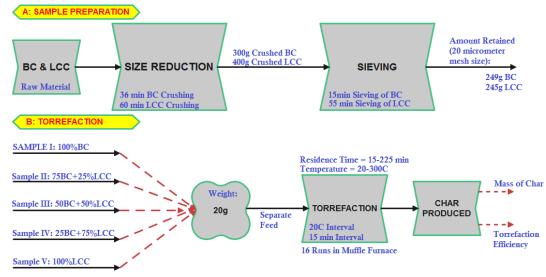


Figure 1. Entire study framework.

Minimization of energy consumption, specialized application and greenhouse gas emission reduction were some of the benefits derived from the LCC and BC torrefaction at < 100 °C, specified in Figure 1. Improvement in energy density, stability and other properties of the torrefied biomass is expected at  $\geq 200$  °C, resulting in char of desired property for industrial usage.

#### 3. Results and Discussion

#### 3.1 Sample Amount After Torrefaction

Sample I at 300 °C, residing for 225 min in the furnace, 7.91 g shiny blackish color of residual char was obtained. The coal char produced has a greater amount of fixed carbon. At 280 °C and a residence time of 210 min, Sample V produced 6.83 g of LCC char, and as the temperature and residence time further increased to 300 °C and 225 min, respectively, a blackish-colored char weighing 6.7 g appeared. Here, greater weight loss occurs compared to BC with a difference of 1.21 g. Clearly, the LCC biomass has lower fixed carbon compared to BC as shown in Table 2.

**Table 2.** Mass of char produced from burning each samples.

T (0C)	Residence Time (min)	Weight of Char (g)				
Temperature (°C)		Sample I	Sample II	Sample III	Sample IV	Sample V
0	0	20	20	20	20	20
20	15	19.13	19.31	19.45	19.57	19.78
40	30	18.73	18.73	18.69	19.14	18.00
60	45	17.53	17.00	18.39	18.40	16.82
80	60	16.66	15.13	17.02	16.71	12.66
100	75	16.03	14.72	15.35	15.03	10.72
120	90	15.71	13.99	15.03	14.79	9.38
140	105	14.53	12.99	14.11	13.55	8.50
160	120	13.72	12.02	13.57	12.73	8.14
180	135	12.11	11.73	12.98	12.38	7.69
200	150	11.53	10.11	12.15	11.10	7.44
220	165	11.07	9.78	10.18	9.33	7.31
240	180	10.53	9.09	8.75	8.69	7.13
260	195	9.12	8.89	7.82	8.00	6.93
280	210	8.72	8.41	7.29	7.81	6.83
300	225	7.91	8.01	6.90	6.82	6.70

Table 2 show the mass of char produced from burning each sample at different temperatures and residence times. It indicates that as the temperature and residence time increase, the mass of char produced decreases for all samples. Simply because the torrefaction process involves heating the samples in a low-oxygen environment, which causes the release of volatile matter and moisture, resulting in weight loss. Sample V, which is 100% LCC, may be suitable for cofiring with coal char due to its lower fixed carbon content and potential for reducing greenhouse gas emissions. On the other hand, the other samples that contain BC may have higher energy density and improved combustion characteristics after torrefaction, making them more efficient for thermal power plants, filtration and metallurgical processes. The higher energy density after torrefaction is due to the removal of moisture and volatile components, which increases the carbon content and calorific value of BC, thereby enhancing its combustion efficiency for thermal and industrial applications. Figures 2 and 3 show the effect of temperature and residence time during the torrefaction process. Based on the information provided in Figure 2, the best property in terms of mass of char obtained (or the optimal torrefaction condition) is at a temperature of 300 °C for Sample I (100% BC). At this temperature and a residence time of 225 min in the muffle furnace, Sample I produced 7.91 g of shiny blackish color residual char, indicating favorable properties for this specific sample. These optimal values are almost in consonance with Tuly et al. [21], who obtained 300 °C and 40 min for bamboo and sub-bituminous coal blends.

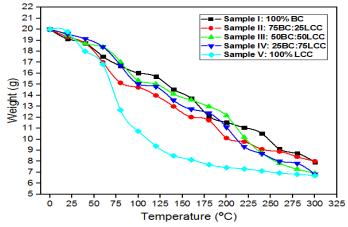


Figure 2. Mass of char against retort furnace temperature for materials and blends.

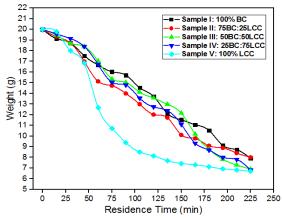


Figure 3. Mass of char against residence time of materials and blends in muffle furnace.

Following Sample I, the next best properties in terms of mass of char obtained were for Sample II (75% BC and 25% LCC biomass) and Sample III (50% BC and 50% LCC biomass) at a temperature of 280 °C and a residence time of 210 min in the muffle furnace. At this temperature and residence time, Sample II produced 8.41 g of char, while Sample III produced 7.29 g of char. For Sample IV (25% BC and 75% LCC biomass) and Sample V (100% LCC biomass), the best properties in terms of mass of char obtained were at a temperature of 260 °C and a residence time of 195 min in the muffle furnace. At this temperature and residence time, Sample IV produced 8.00 g of char, while Sample V produced 6.93 g of char. But Ajikashile et al. [13]'s findings show that 240 °C torrefaction temperature and 60 min residence time are the optimal torrefaction conditions for millet and sorghum straw torrefaction.

Residence time refers to the duration that the sample is exposed to the torrefaction process. According to Figure 3, the residence time has a significant effect on the mass of char produced from torrefaction. As the residence time increase, the mass of char produced decreases for all samples. Also, the rate of weight loss varies for different samples. For example, Sample I, which is 100% BC, has a higher rate of weight loss compared to the other samples. This is because BC has a higher volatile matter content compared to LCC biomass, which is the primary component of the other samples. As a result, BC releases more volatile matter and moisture during torrefaction, leading to a higher rate of weight loss. Furthermore, the figure show that the weight loss is affected by the composition of the sample. For example, Sample II, which is a blend of 75% BC and 25% LCC biomass, has a lower rate of weight loss compared to Sample I, but a higher rate of weight loss compared to Sample V, which is 100% LCC biomass. This indicates that the presence of LCC biomass in the sample can reduce the rate of weight loss during torrefaction.

#### 3.2 Char Produced

All five samples changed their color after heat treatment, as observed. Figure 4 indicates that the char produced from the torrefaction of BC (Sample I) has a shiny blackish color, while the char produced from the torrefaction of LCC biomass (Sample V) has a brownish color. The char produced from the other samples (blends of BC and LCC biomass) exhibit varying shades of black and brown, depending on the composition of the sample. At 300 °C, coal undergoes mild torrefaction (or low-temperature devolatilization), which typically causes slight weight loss and structural changes but does not fully carbonize the material. The resulting char is usually brittle and may partially crumble into fine particles, though it won't be completely powdery, as shown in Figure 4. It retains some solid structure unless ground or handled roughly afterward.

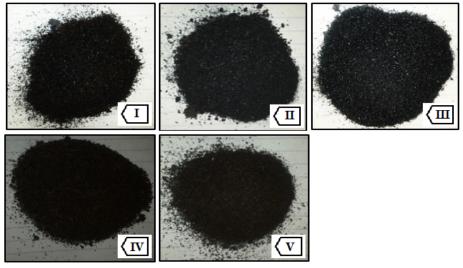


Figure 4. Char after heat treatment at 300 °C and 30 min.

The color of the char produced after torrefaction is a crucial indicator of its properties. A shiny blackish color indicates that the char has a high carbon content and low volatile matter content, which are desirable properties for combustion. On the other hand, a brownish color indicates that the char has a lower carbon content and higher volatile matter content, which may result in lower combustion efficiency. Therefore, the findings from Figure 4 suggest that the composition of the sample can affect the properties of the char produced after torrefaction. Blends of BC and LCC biomass may result in char with intermediate properties, depending on the ratio of the two components. Potential biochar applications include pollution remediation, soil fertility improvement, and carbon sequestration [22,23]. But researchers must examine the different type of colors presented and their specific applications in relevant industry.

## 3.3 Impact of Mass Yield

Torrefaction efficiency can be expressed in form of mass yield. Solid or mass yield is a measure of the solid yield of the torrefaction process or the fraction of the original

component of biomass that is transformed into solid char, as defined by Preradovic et al. [20] and shown in Table 3.

<b>Table 3.</b> Calculated coal and biochar mass yield	Table 3.	Calculated	coal and	biochar	mass	vields.
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D	M (-)	Mass Yield (y <sub>M</sub> )					
Run	$M_{feed}$ (g)	Sample I	Sample II	Sample III	Sample IV	Sample V	
1.	20	1	1	1	1	1	
2.	20	0.9565	0.9655	0.9725	0.9785	0.989	
3.	20	0.9365	0.9365	0.9345	0.957	0.9	
4.	20	0.8765	0.85	0.9195	0.92	0.841	
5.	20	0.833	0.7565	0.851	0.8355	0.633	
6.	20	0.8015	0.736	0.7675	0.7515	0.536	
7.	20	0.7855	0.6995	0.7515	0.7395	0.469	
8.	20	0.7265	0.6495	0.7055	0.6775	0.425	
9.	20	0.686	0.601	0.6785	0.6365	0.407	
10.	20	0.6055	0.5865	0.649	0.619	0.3845	
11.	20	0.5765	0.5055	0.6075	0.555	0.372	
12.	20	0.5535	0.489	0.509	0.4665	0.3655	
13.	20	0.5265	0.4545	0.4375	0.4345	0.3565	
14.	20	0.456	0.4445	0.391	0.4	0.3465	
15.	20	0.436	0.4205	0.3645	0.3905	0.3415	
16.	20	0.3955	0.4005	0.345	0.341	0.335	

Equation 1 based on  $M_{\rm feed}$ = 20g and obtained  $M_{\rm Char}$  in Table 2 gives the torrefaction efficiency or  $y_{\rm M}$  of the respective sample (Table 3) at various residence time and temperature. At 20 °C,  $y_{\rm M}$  ranges from 95.65-98.9% during the 5 samples first run in muffle furnace to obtain their char. The slight net mass loss (100 –  $y_{\rm M}$ ) observed at 20 °C (4.35-1.10%) is likely due to minor moisture evaporation, rather than any thermal decomposition since the temperature is too low to induce significant chemical changes. A progressive decrease in efficiency is observed over the entire run, up to the smallest  $y_{\rm M}$  at the highest temperature set, as shown in Figure 5. Because, at higher temperatures, volatile components such as water, hemicellulose and some volatile organic compounds are driven off, leading to a reduction in the overall mass of the biomass. The remaining material is rich in carbon and has an increased energy density, making it suitable for various applications such as bioenergy production.

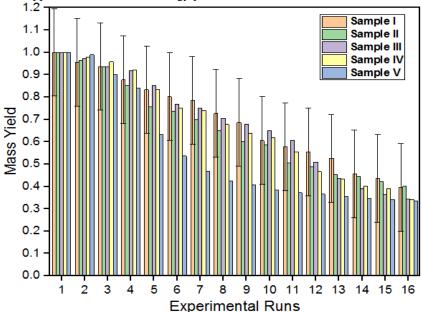


Figure 5. Mass yield of each sample within the stated temperature range.

Some other applications of torrefied biomass are as high-quality fuel for innovative bioenergy application, biomass solid fuel for thermal power plants for generation of heat and electricity, co-gasification in entrained-flow gasifier, pellets or briquettes as fuels, fuels for domestic and commercial use, small-scale pellet boilers or stoves and cofiring in pulverized boilers [20]. Preference for high or low mass yield during torrefaction depends on the specific application and goals. If the goal is to maximize the yield of torrefied biomass (Samples I-V), choosing conditions that result in a higher  $y_M$  can be preferable, which means more material is retained after torrefaction. However, high  $y_M$  may come at the expense of energy density, as a significant portion of the biomass may still contain volatile components. If the focus is on maximizing the energy density of the torrefied biomass, opting for conditions that result in a lower  $y_M$  (i.e., at 33.5-40% for the samples) could be more suitable, with the lowest being Sample V with  $y_M$ = 0.335. This typically involves higher temperatures that remove more volatile components, according to Khairy et al. [24]. The drawbacks in this situation are that less material is retained, and the process may be less efficient in terms of biomass utilization.

Error bars in bar charts, based on standard deviations, provide a visual representation of the variability or uncertainty associated with each data point. The narrowness of the error bars in Figure 5 suggests greater precision and reproducibility, while wider error bars indicate more variability. As observed, percent of data error decreased from 0.05-0.01978 in Run 1-16, corresponding to a standard deviation of 0.19607. If a data point is 0.05 and the associated error bar is 0.19607, it might suggest that the data point has a relatively large percentage uncertainty compared to the value itself. But, at lower mass yield, the uncertainty diminishes towards the sample with more reasonable mass yield.

#### 4. Conclusion

Separate torrefaction of 100% BC, 100% LCC and 3:1, 1:1 & 1:3 blends was effectively carried out using a Retort furnace, resulting in the production of char of dissimilar weight and color with unique feasibility for industrial use. It is well known that torrefaction may potentially produce a solid fuel with combustion reactivity and porosity equivalent to raw biomass while having compatible energy density and grindability to coal. Sample V (which is purely LCC) best suits a co-firing bioenergy plant as it has the lowest fixed carbon content, lowest weight loss (13.3 g) and higher volatile matter content with a brownish color char of low mass yield (13.3%) and maximum energy density at 300 °C and 225 min residence period. Notwithstanding, the blends (Sample II, III & IV) char can go into several industrial usage explained earlier above the property demonstrated by 100% BC. Ongoing and future research should focus on optimizing torrefaction conditions for specific feedstocks, the exploration of different torrefaction technologies and assessing the performance of torrefied materials in industrial settings. In line with that, kinetic and thermodynamic parameters, enhancement factor, energy yield, torrefaction severity index, fuel ratio, decarbonization, dehydrogenation and deoxygenation rates must be determined and analyzed. Future works should look at minimum elemental constituents, volatile and ash content, thermogravimetric analysis (TGA) and high heating value (HHV) contents of the biomass or char, which is the main drawbacks of the study herein.

#### **Conflicts of Interests**

The authors declare they have no conflicts of interests.

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#### **Data and Materials Availability**

All data associated with this study are present in the paper.

#### **Generative AI Statement**

The authors declare that no Gen AI was used in the creation of this manuscript.

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