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To cite this article: Y T Lee *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **354** 012074

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Thermochemical upgrading of coconut husk and rubber seed to coal co-firing feedstock via torrefaction

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Abstract. This research seeks to investigate thermochemical upgrading of Malaysian coconut husk and rubber seed to coal co-firing feedstock via torrefaction. Torrefaction experiments within a range of temperatures (200-300°C) and holding times (30 and 60 min) were carried out using a tubular furnace. Mass yield, higher heating value (HHV), energy densification ratio (EDR) and energy yield as well as proximate analysis were determined for the torrefied biomass sample. Both torrefaction temperature and holding time decreased the mass yield and the energy yield of the torrefied biomass. The HHV and EDR of coconut husk and rubber seed increased upon torrefaction. The VM decreased while the FC increased after torrefaction. Rubber seed torrefied at 300°C and 60 min had a HHV of 29.22 MJ/kg which was comparable to that of bituminous coal. Linear correlations ($R^2=0.7764-0.965$) were developed between mass yield and EDR, as well as mass yield and energy yield. Up to 70 wt% and 60 wt% of coal with a fuel ratio of 1 could be replaced by coconut husk and rubber seed torrefied at 300°C and 60 min, respectively. Overall, the study indicated the feasibility of torrefied rubber seed (300°C and 60 min) as a coal co-firing feedstock in comparison to torrefied coconut husk due to its higher HHV cum lower ash content as well as its high co-firing proportion with coals having fuel ratio of at least 1.

1. Introduction

In light of global warming due to increasing atmospheric carbon dioxide linked to fossil fuels use, it is both crucial and timely to diversify our energy sources and reduce reliance on fossil fuels. Biomass is an alternative renewable energy source. Presently, biofuels cum biomass constitute only 9.7% of the world total primary energy supply. There is a good potential for more utilization of this source considering the vast availability of biomass, estimated at 500 to 560 billion tonnes of carbon [1]. Nonetheless, there are challenges with regard to the application of biomass as a fuel in present day coal power stations, mainly due to the physicochemical properties of the biomass. Compared to coal, biomass contains more moisture and has lower heating value in addition to being more inhomogeneous [2]. These lead to difficulties in combustion and issues with transportation and storage. Torrefaction is a thermochemical upgrading process which has been proposed to enhance the fuel properties of biomass. It is a mild pyrolysis process whereby the feedstock is heated between temperatures of 200 °C to 300 °C. The resulting torrefied biomass has higher energy density, more



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homogeneous composition and improved grindability [3].

Within the Malaysian context, the choice of biomass as feedstock for torrefaction is vast considering the varied agriculture and tropical diversity. In this research, two types of biomass i.e. coconut husk and rubber seed have been selected as the torrefaction feedstock. Coconut and rubber are major crops in Malaysia with an estimated production of 504.8 and 673.5 MMT in 2016 [4,5]. Coconut husk is considered as an agricultural waste. Rubber seed, in contrast, could be used for the germination of rubber trees, but is generally considered as waste [6]. This main aim of this research is to investigate and compare the viability of upgrading these agricultural wastes to be a coal co-firing feedstock via torrefaction.

2. Materials and methods

2.1. Materials

Coconut husk and rubber seed were obtained from a wet market and a rubber plantation in the state of Selangor Darul Ehsan in Malaysia, respectively. The biomass samples were air dried for a week to establish a basis for the experiments. The dried samples were then ground and sieved to 0.5 mm using a Retsch ZM200 centrifuge grinder, and stored in the absence of light in airtight containers until torrefaction was carried out.

2.2. Torrefaction runs

The torrefaction runs were carried out in a Carbolite CTF 12/65/550 ceramic tubular furnace with an inner diameter of 65 mm and a heated length of 550 mm. Ground biomass of 20 g weight was placed onto a ceramic boat. Then, the ceramic boat was placed in the middle of the tubular furnace and its ends were sealed. Nitrogen (N₂) gas was flowed through the furnace for 5 min at 40 cm³/min to purge inert gases. Subsequently, the furnace was switched on and its temperature was raised to levels of 200, 250 or 300 °C, at a heating rate of 10 °C/min. For each temperature level, two holding times (30 and 60 min) were investigated. The N₂ flow rate was maintained at 40 cm³/min for all runs. After the specified holding time, the furnace was switched off and cooled to ambient temperature. The torrefied biomass sample was weighed at ambient temperature and stored in an airtight container for further analyses. Each experimental run was carried out twice and the average values were obtained. The mass yield for each torrefaction run was determined from equation (1):

$$\text{Mass yield (\%)} = \frac{\text{Final mass (g)}}{\text{Initial mass (g)}} \times 100\% \quad (1)$$

2.3. Higher heating value (HHV)

The higher heating value (HHV) of the raw and torrefied biomass samples was experimentally determined using a Parr 6100 Bomb Calorimeter. The energy densification ratio (EDR) and energy yield for the torrefied biomass samples were then calculated from the HHV of the raw and torrefied biomass samples following equation (2) and (3), respectively.

$$\text{EDR} = \frac{\text{HHV of torrefied sample (MJ/kg)}}{\text{HHV of raw sample (MJ/kg)}} \quad (2)$$

$$\text{Energy yield (\%)} = \text{EDR} \times \text{Mass yield (\%)} \quad (3)$$

2.4. Thermogravimetric analysis (TGA)

Thermogravimetric analysis (TGA) was used to carry out proximate analyses of the raw and torrefied biomass samples at the most severe torrefaction conditions of 300°C and 60 min using a Mettler Toledo TGA/DSC 1. For proximate analysis, an appropriate TGA temperature profile was established first by referring to British Standards BS EN 18134 (moisture content determination), BS EN 15148

(volatile matter (VM) content determination) and BS EN 14775 (ash content determination). At the fixed 50 mL/min N₂ flow rate, the sample was heated from 30°C at 10 °C/min till 105 °C and maintained at 105 °C for 40 min. The mass loss which occurred in this segment represented the moisture content of the sample. Subsequently, at 20 °C/min, the temperature was raised to 900 °C, maintained isothermally for 7 min and then decreased to 550 °C at the same rate. The mass loss in this region indicated the VM content of the sample. At 550 °C, the gas was swapped to oxygen gas flowing at 50 mL/min and this temperature was constant for 2 h. The remaining sample mass at the end of the analysis was the ash content. The fixed carbon (FC) content percentage was determined from the difference between 100 and the total sum (%) of moisture, VM and ash.

3. Results and discussion

3.1. Mass yield, HHV, EDR and energy yield

Table 1 summarises the determined mass yield, HHV, EDR and energy yield for all the tested torrefaction conditions. It could be clearly observed that the mass yield reduced with increasing torrefaction temperature and holding time. This was attribute to the loss of moisture and volatile matter from the biomass during the thermochemical process [7,8]. Taking coconut husk as an example, when the temperature increased from 200 °C to 300 °C, the mass yield decreased by 15.75% and 26% for 30 and 60 min holding times, respectively. When the holding time was increased from 30 min to 60 min, reductions of 3.5%, 9.1% and 13.75% were recorded for temperatures of 200 °C, 250 °C and 300 °C, respectively. Thus, it could be seen that the increase in temperature affected the mass yield more significantly as compared to the increase in holding time. A similar trend was observed for rubber seed. Under the most intense torrefaction conditions of 300°C and 60 min, the mass yield of torrefied rubber seed was lower than that of torrefied coconut husk.

As shown too in table 1, increasing the torrefaction temperature resulted in increased HHV of the torrefied biomass. The raw coconut husk had a HHV of 16.747 MJ/kg, which was comparable to that reported in the literature [9]. Raising the torrefaction temperature from 200 °C to 300 °C resulted in the HHV increasing from 17.206 MJ/kg to 20.206 MJ/kg and from 17.622 MJ/kg to 22.149 MJ/kg for 30 min and 60 min holding times, respectively. Correspondingly, the calculated EDRs increased from 1.027 to 1.207 (18%) and from 1.052 to 1.323 (27.1%). From the definition of EDR given in Eqn. [2], the values represented the increase in the biomass energy density. Likewise to the effect of holding time on mass yield, increasing the holding time had a less significant effect on the HHV and thus EDR. Prolonging the holding time from 30 min to 60 min caused the EDR to increase by 2.5%, 2.0% and 11.6% for temperatures of 200 °C, 250 °C and 300 °C, respectively. Meanwhile, the HHV of rubber seed was 24.954 MJ/kg, a value close to the reported values by [10]. Again, a similar trend in EDR with torrefaction severity could be observed for rubber seed.

Table 1. Mass yield, HHV, EDR and energy yield of raw and torrefied coconut husk and rubber seed samples.

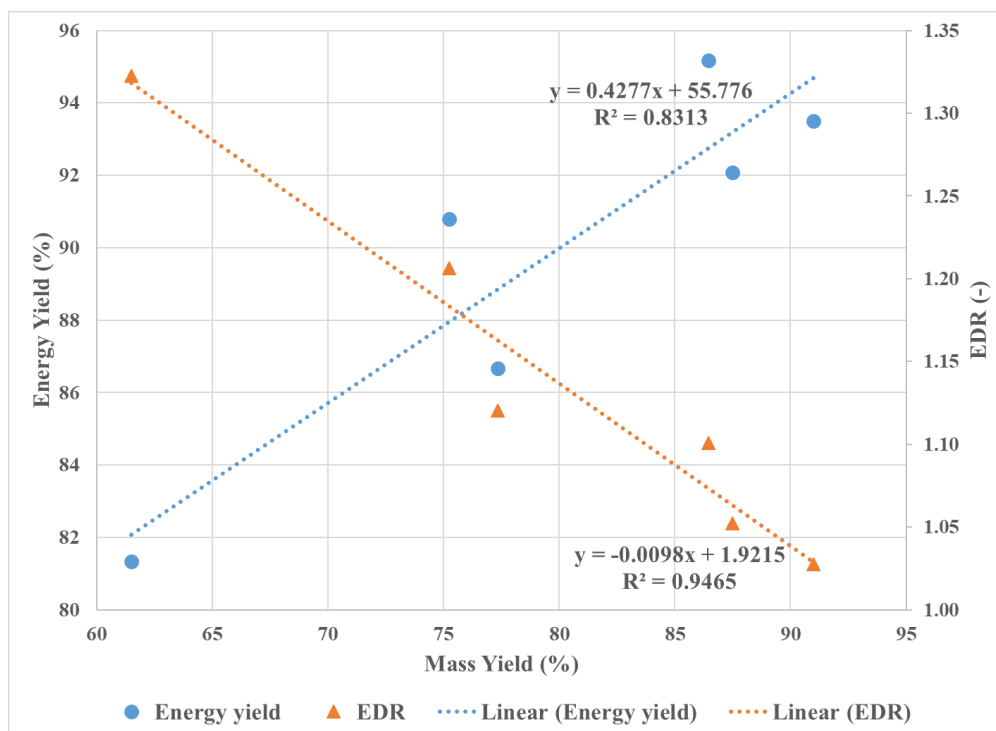
Biomass	Parameter	Raw	200°C 30 min	200°C 60 min	250°C 30 min	250°C 60 min	300°C 30 min	300°C 60 min
Coconut husk	Mass yield (%)	-	91.000	87.500	86.450	77.350	75.250	61.500
	HHV (MJ/kg)	16.747	17.206	17.622	18.437	18.766	20.206	22.149
	EDR (-)	-	1.027	1.052	1.101	1.121	1.207	1.323
	Energy yield (%)	-	93.494	92.072	95.174	86.675	90.792	81.338
Rubber seed	Mass yield (%)	-	96.750	92.350	83.600	68.400	77.900	57.750
	HHV (MJ/kg)	24.954	25.098	25.175	26.173	26.417	27.712	29.220
	EDR (-)	-	1.006	1.009	1.049	1.059	1.111	1.171
	Energy yield (%)	-	97.308	93.168	87.684	72.410	86.510	67.623

The obtained trends in HHV and EDR can be explained by the underlying processes occurring during torrefaction. Moisture loss from the biomass during torrefaction results in a decrease of hydrogen and oxygen without loss of carbon. During torrefaction too, VM which typically consists of compounds of hydrogen and oxygen as well as light hydrocarbons with low carbon content relative to hydrogen [7] is lost. Hence, a significantly higher amount of hydrogen and oxygen are lost during torrefaction in comparison to carbon [8]. Due to the reduction in the H:C and O:C ratios and the fact that C-H and C-O bond energies are lower than that of C-C, the HHV of the torrefied biomass increases as a result [11].

As can be seen in equation [3], the energy yield is a function of the EDR and the mass yield. With higher torrefaction temperature or longer holding time, the EDR increased as explained earlier and thus, this increased the energy yield. Conversely, the mass yield decreased with increasing torrefaction temperature or holding time, and this resulted in a negative effect on the energy yield. The generally decreasing trend in energy yields with increasing torrefaction temperature or holding time as observed in table 1 was because the increase in the EDR was insufficient to compensate for the decrease in the mass yield.

3.2. Correlations between mass yield, EDR and Energy yield

Figure 1 illustrates the linear correlations that were developed between mass yield and EDR as well as mass yield and energy yield. Coconut husk displayed a strong correlation between mass yield and EDR, with an R^2 value of 0.9465. As for the correlation between mass yield and energy yield, a reasonably high R^2 value of 0.8313 was obtained for coconut husk. The linear correlations noted here agreed with the findings of Álvarez *et al* [12]. In comparison to coconut husk, rubber seed showed a strong correlation between mass yield and energy yield ($R^2=0.965$), but had a weaker correlation between mass yield and EDR ($R^2=0.7764$). One possible reason for the observed trend in rubber seed in comparison to coconut husk and other biomass reported in the literature [12] could be that it is not a lignocellulosic-rich feedstock. Instead, rubber seed contains protein (22.51%) and fat (50.91%) as reported in [6] thus this suggests that it may have different thermochemical behaviour under torrefaction.



(a)

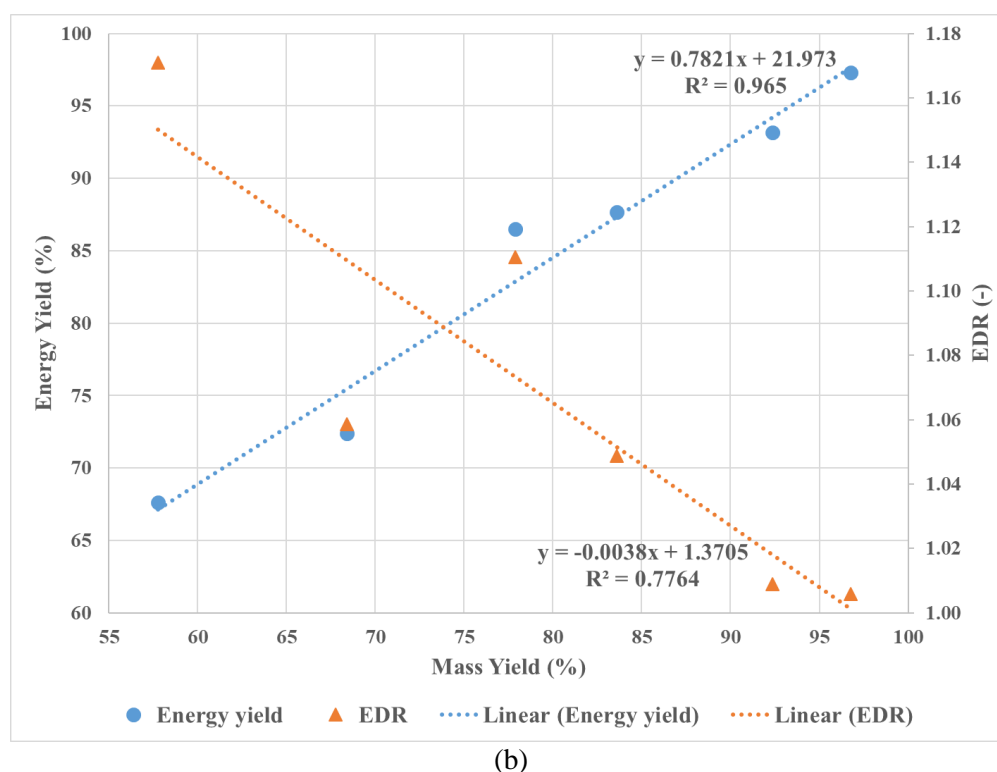


Figure 1. Correlations between for mass yield and EDR as well as mass yield and energy yield for coconut husk (a) and rubber seed (b).

3.3. Proximate analysis

On a dry basis, the proximate analyses of the raw coconut husk and rubber seed samples were 73.381% VM, 22.953% FC and 4.667% ash for the former, and 87.869% VM, 8.947% FC and 3.184% ash for the latter. The results for raw coconut husk compare well with that obtained by Said *et al* [9]. For raw rubber seed, the results are within the ranges obtained by other researchers [10,13]. The VM content of coconut husk decreased by 8.551% on a dry basis after torrefaction. This was accompanied by an increase of 0.345% in FC content from 22.953% to 23.298% on a dry basis. Similarly, on a dry basis, rubber seed had a decrease of 7.906% for VM content and an increase of 8.249% for FC content. The loss of VM corresponded to the higher HHV and EDR as explained earlier in Section 3.1. The increase in FC is one of the drivers for carrying out torrefaction as an upgrading process. Despite the seemingly low percentage increase in FC content for coconut husk, its FC content (dry) after torrefaction was actually higher than that of torrefied rubber seed at 23.298%. Finally, the ash content increased for coconut husk and decreased slightly for rubber seed after torrefaction was carried out. The final dry ash content of coconut husk at 12.872% was considered high. The use of biomass with high ash content has been linked to an increase in slagging propensity [14]. Therefore, a low ratio of co-firing with coal would seem appropriate in order to reduce the slagging effect.

3.4. Comparison of HHV and EDR with other torrefied biomass

Table 2 compares the HHV and EDR of the torrefied biomass in this study with other torrefied biomass reported in the literature [15-18]. To ensure a basis for comparison, all the listed values were measured at the most intense torrefaction conditions of 300°C and 60 min. As can be seen in table 2, the HHV of raw biomass ranged from as small as 12.66 MJ/kg to as much as 24.954 MJ/kg. After torrefaction, enhancement of HHV or EDR in percentage varied from 12% to 36.7%. This corresponds to the study by Arnsfeld *et al* [19] who reported that generally, the HHV of raw wastes range between 16 and 21 MJ/kg, which increases to between 18 and 28 MJ/kg after torrefaction. The data in table 3

highlights the range of possible outcomes of torrefaction on different biomass types at the most important torrefaction conditions of temperature and holding time. It should be noted, however, that other torrefaction parameters for instance heating rate and particle size may also impact the compiled results. The EDRs measured for coconut husk and rubber seed were within the range of outcomes reported in the literature. Coconut husk had the higher EDR, comparable to that of Chinese medicine residue [15]. In contrast, rubber seed has a much lower EDR of 1.171 (17.1%), which was comparable to olive tree pruning at 1.12 (12%) [17]. However, the final HHV of rubber seed was much higher than that of coconut despite its lower EDR as shown in table 1 (29.22 MJ/kg versus 22.149 MJ/kg).

Table 2. Comparison of HHV and EDR of different torrefied biomass at 300°C and 60 min.

Biomass	HHV (MJ/kg)	EDR (-)	Reference
Spent ground coffee	21.77	1.367	15
Chinese medicine residue	20.38	1.340	15
Microalgae residue	12.66	1.367	15
Rice husk	15.3	1.242	16
Olive tree pruning	17.32	1.12	17
Oil palm frond	22.55	1.296	18
Coconut husk	16.747	1.323	<i>Present study</i>
Rubber seed	24.954	1.171	<i>Present study</i>

Table 3. Variation of fuel ratio with biomass co-firing proportions with coal having a fuel ratio of 1.

Biomass	Fuel ratio for different biomass co-firing proportions (wt%)										
	0	10	20	30	40	50	60	70	80	90	100
Torrefied coconut husk	1	0.936	0.873	0.810	0.746	0.683	0.619	0.556	0.492	0.429	0.365
Torrefied rubber seed	1	0.922	0.843	0.765	0.686	0.608	0.529	0.451	0.372	0.294	0.215

3.5. Feasibility of co-firing with coal

From the viewpoint of using torrefied biomass for co-firing in coal power plants, the HHV instead of the mass of feedstock is the primary concern. This is because in co-fired power plants, the share of biomass is generally defined by the amount of useful heat from the biomass, which is directly linked to the reduction in greenhouse gas emission per unit MWh generated and thus the carbon credit of the plant [20]. From this study, it could be seen that torrefied rubber seed at 300°C and 60 min had the highest HHV of 29.22 MJ/kg, which was comparable to that of bituminous coal with HHVs between 23 MJ/kg and 26 MJ/kg [21]. This implied that it could be used as a partial replacement for coal for power generation.

In addition to HHV, the fuel ratio of a fuel, defined as the ratio of FC to VM on a dry basis is also important. In coal-fired power plants, fuel ratios are typically within the range of 0.5 to 2 [22]. The fuel ratio of coconut husk and rubber seed increased after torrefaction at 300°C and 60 min, from 0.317 to 0.365, and 0.102 to 0.215, respectively. Nevertheless, the fuel ratios were below the typical minimum thresholds of 0.5. This can be resolved by co-firing the torrefied biomass with coal. Assuming coal with a fuel ratio of 1 is used, table 3 illustrates the relationship between the co-firing proportions for both torrefied biomass and the fuel ratios of the mixed fuel. Here, it can be seen that up to 70 wt% of coal could be replaced by torrefied coconut husk whereas up to 60 wt% of torrefied rubber seed could be used to switch with coal. Both the proportions were considerably high. Bearing in mind the high ash content of torrefied coconut husk, torrefied rubber seed would appear as a better coal co-firing feedstock in comparison to torrefied coconut husk. This is further strengthened by its higher HHV cum lower ash content as well as its high co-firing proportion with coals having fuel ratio of at least 1.

4. Conclusions

Thermochemical upgrading of Malaysian coconut husk and rubber seed to coal co-firing feedstock via torrefaction has been investigated. Increasing the torrefaction temperature and holding time decreased the mass yield and the energy yield of the torrefied biomass albeit with a less pronounced effect for holding time. The HHV and EDR of both biomass samples increased upon torrefaction, with rubber seed torrefied at 300°C and 60 min having a HHV of 29.22 MJ/kg which was comparable to that of bituminous coal. The HHV and EDR of torrefied coconut husk and rubber seed were also comparable to other torrefied biomass reported in the literature. Linear correlations ($R^2=0.7764-0.965$) were developed between mass yield and EDR, as well as mass yield and energy yield. After torrefaction, the VM of the biomass decreased while the FC and ash contents increased. Up to 70 wt% and 60 wt% of coal with a fuel ratio of 1 could be replaced by coconut husk and rubber seed torrefied at 300°C and 60 min, respectively. Overall, torrefied rubber seed (300°C and 60 min) was deemed to be a more suitable coal co-firing feedstock due to its higher HHV cum lower ash content as well as its high co-firing proportion with coals having fuel ratio of at least 1.

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