Discussion on Torrefaction Parameters for Making the Refuse-Derived Fuels by Response Surface Methodology

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Abstract. The torrefaction process is one of the mild pyrolysisor the low-temperature carbonization, which is a thermochemical conversion in which biomass is heated in an inert or nitrogen atmosphere, and the temperature ranges from 200°C to 300°C. It is considered to be an effective pretreatment method to make solid fuels. This study utilizes low-temperature carbonization technology to improve the combustion efficiency of solid-derived fuel (RDF-5) made from biomass. In this experiment, a tubular furnace was used to feed nitrogen gas for 15 min, and four raw materials of pennisetum, rice straw, wood chips and Camellia seed cakes were fed. The analysis was carried out, and the effects of temperature and residence time on the calorific value, energy yield and energy density of the four raw materials were explored using response surface methodology (RSM). It can be seen from the experimental results that among the four raw materials, the carbon content and calorific value of Camellia seed cakes are the highest. Elemental analysis showed that the proportions of nitrogen and sulfur in these four raw materials are very low. These four substances were understood to be suitable to be refuse-derived fuels (RDF-5). Comparing the calorific value of the four kinds of biomass after torrefaction process, the calorific value of pennisetum, Camellia seed cakes, wood chips and rice straw increased by 42.5%, 38.7%, 52.7%, and 18.6%, respectively. These findings revealed that the higher temperature and the longer residence time, the smaller energy yield and the higher energy density.

1 Introduction

Taiwan's annual agricultural waste is about 4.5 million metric tons, and rural areas are often burned on site, causing air pollution and environmental problems. According to statistics from the Energy Bureau of the Ministry of Economic Affairs, the proportion of Taiwan's self-produced energy in 2020 will increase from 2.1% to 2.2%, but there are still 97.8% of the energy supply needs to be imported. With the rising awareness of environmental protection and clean energy, if agricultural and forestry wastes can be made into biomass energy, it will not only solve the problem of agricultural wastes, but also convert the use of energy will get the most out of it. Nowadays, countries such as the European Union, China and Southeast Asia are also actively researching and developing refuse-derived fuel (RDF-5). Compared with fossil fuels, RDF-5 has the characteristics of low sulphur and carbon neutral. The RDF-5 can be produced through pretreatments such as drying, crushing, mixing and granulation.

The raw materials for the manufacture of RDF-5 can be divided into five categories, namely agricultural waste (sugar cane, corn, sugar beet, soybean, etc.), forestry waste (wood, forestry waste, etc.), industrial organic waste (organic sludge, waste plastics, etc.), animal husbandry waste (animal manure, animal carcasses, etc.) and municipal solid waste (domestic waste). RDF-5 mainly homogenizes these wastes through the first crushing after recycling the waste. At the same time, the step of magnetic separation is carried out using electromagnets to select metal substances, and then the waste is dried by hot air to reduce the moisture content. Secondary crushing, the waste is evenly crushed to facilitate its granulation, and finally additives are added to extrusion moulding. Granulation can increase the fuel density and make it into a uniform shape, which is convenient for its storage and transportation. The calorific value of RDF-5 is generally about 3500-4500 kcal/kg, which is two-thirds of the calorific value of the coal. Thus, RDF-5 can be directly put into the boiler as fuel or mixed with coal, and has the most vigorous application.

Torrefaction process is a low-temperature carbonization and thermochemical conversion in which biomass is heated in an inert or nitrogen atmosphere, and the temperature ranges from 200°C to 300°C. It is considered to be an effective thermal pretreatment method for producing biomass energy [1-3]. The biomass torrefaction process can be mainly divided into four stages, including water evaporation, hemicellulose decomposition, lignin decomposition and cellulose decomposition [3]. Five basic components (hemicellulose, cellulose, lignin, xylan, and dextran) and two pure materials (xylose and glucose) were torrefied to study the thermal degradation properties of the test samples.

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Torrefaction at 230°C released only some moisture and light volatiles from the test samples, which had relatively little effect on improving the biomass properties. Torrefaction at 260°C resulted in a certain amount of pyrolysis of hemicellulose, while cellulose and lignin were barely affected. It is expected thatthe properties and calorific value of the torrefied biomass can be enhanced to a certain extent. When biomass was torrefied at 290 °C, a large amount of hemicellulose and cellulose were destroyed [4]. Below 500 °C, the pyrolysis of hemicellulose and lignin involves exothermic reactions, while the pyrolysis of cellulose is endothermic. No organic compounds were detected in lignin pyrolysis, and more C=O organic compounds appeared in hemicellulose, while cellulose had higher -OH and C-O contents, and more $-O-CH_3$ [5].

Generally, in pyrolysis and low-temperature carbonization, both temperature and heating rate increase the rate of thermal degradation, resulting in more liquid products instead of solid products. The increase in temperature reduces the yield of carbonization, and the yield decreases exponentially above 250 °C. For reactive polymers during torrefaction, the residence time mainly affects the degradation of hemicellulose, while cellulose has limited weight loss at short reaction times (less than 30 min). Temperature mainly affects the occurrence and kinetics of decomposition reactions, and it is proposed that a small increase in temperature increases the degradation rate, while a significant increase in reaction time; an increase in residence time favours gas formation [6].

During torrefaction process, solid, liquid, and gaseous products will be produced, and the output depends on temperature and residence time in the furnace. The solid reaction product has a complex structure, including a modified sugar structure, a newly formed polymer structure that may have an aromatic ring, and a typical carbon-rich structure and ash content. The gas product is relatively fixed, mainly CO_2 and CO, followed by a small amount of CH_4 and H_2. Other compounds contain light aromatic components, such as benzene and toluene, and their boiling point is lower than -33 °C. Its liquid products, which are not the main reaction products, contains compounds such as waxes and fatty acids, dehydrosugars from cellulose, furfural and acetic acid from hemicellulose, and phenols from lignin, and inert products may be torrefied. In the case of being evaporated, some of its liquid products can be solid at room temperature [6,7].

In this study, pennisetum, wood chips, rice straw and press cake of camellia seeds were collected to evaluate the differences of four raw materials (herbaceous plants, oil crops and woody biomass). In the preliminary detections of the characteristics of raw materials to understand the basic properties, torrefaction was conducted in a tubular furnaces and oxygen-free conditions at different temperatures and different residence times. Different raw materials have different responses on temperature and residence time. As the results, calorific value, energy density, and energy yield were optimized by response surface methodology (RSM), and the calorific value and combustion properties of the species after the low-temperature carbonization were explored using the thermal calorimeter and thermo-gravimetric analyser. The present study will help to establish an effective method for designing torrefaction process for the evaluation and utilization of agricultural and forestry waste in the future, which will alleviate waste treatment problems and reducing environmental pollution.

2 Experimental Section

In this study, pennisetum, press cake of camellia seeds, wood chips, and rice straw were used as raw materials, and calorific value analysis of the four raw materials were initially carried out to explore the feasibility of using them as the solid fuels. The relative water content of the four raw materials is less than 15%. Torrefaction experiments in a tubular furnace to explore the influence of various operating parameters on the responses of calorific value, energy density, and energy yield under different parameters of temperatures and residence times were carried out. Response surface Methodology (RSM), were used to explore the temperature (X_1) and residence time (X_2) of the four raw materials to find the torrefaction process responses of calorific value (Y_1), energy yield (Y_2) and energy density (Y_3).

3 Results and Discussion

3.1 Calorific value analysis

The calorific value is analysed using calorie meter (IKA C200). Among the four raw materials, the press cake of camellia seeds has the highest calorific value (5045 kcal/kg), followed by wood chips (4148 kcal/kg) and pennisetum (4079 kcal/kg), rice straw (3885 kcal/kg). There is not much difference between pennisetum calorific value and pennisetum whole plant calorific value (3891 kcal/kg) [8]. Through elemental analysis, it can be known that the heat quality of biomass is positively correlated with its carbon and hydrogen elements, because elemental carbon and hydrogen are the main sources of heat generated during biomass combustion [9].

3.2 Experimental design by response surface methodology

In this study, the torrefaction experiments were carried out in an electrical heating tubular furnace. Through the discussion of the aforementioned operating parameters, using pennisetum, wood chips, press cake of oil seeds and rice straw as raw materials, it can be found that the temperature (X_1) and residence time (X_2) have a certain influence on the calorific value. Therefore, in this study, different temperatures of 210, 250, and 290°C were used for low-temperature carbonization, and the residence times of wood pellets and rice straw were 20, 40, and 60 min, and pennisetum and press cake of seeds were used for different residence times of 20, 50, and 80 min. Under the influence, the RSM experiment is used to evaluate the relationship between the two factors, and to discuss the responses of calorific value (Y_1), energy density (Y_2) and energy yield (Y_3).

The calorific value (Y_1), energy density (Y_2) and energy yield (Y_3) of pennisetum, Camellia seed cakes, wood pellets, and rice straw were shown in Tables 1, 2, 3 and 4, respectively.

Table 1. Experimental data of pennisetum.

NO.	Pattern	X_1 (min)	X_2 (°C)	Y_1 (kcal/kg)	Y_2 (%)	Y_3 (%)
1	0A	50	290	5737	46.22	1.42
2	+−	80	210	5651	50.18	1.39
3	a0	20	250	5794	55.18	1.42
4	++	80	290	5747	53.84	1.41
5	00	50	250	5311	57.54	1.30
6	−+	20	290	5651	50.18	1.39
7	0	50	250	5075	61.96	1.24
8	A0	80	250	5398	55.2	1.17
9	0a	50	210	4725	37.8	1.16
10	−−	20	210	5496	54.82	1.35

According to the experimental data, after pennisetum is torrefied, the best calorific value (Y_1) appears in the third group of experimental conditions, the best energy density (Y_2) appears in the seventh group of experiments, and the best energy yield (Y_3) appears in the first and third groups of experiments. If the third group of operating conditions (20min and 250°C) is selected, Y_1, Y_2 and Y_3 have acceptable values.

Table 2. Experimental data of press cake of camellia seeds.

NO.	Pattern	X_1 (min)	X_2 (°C)	Y_1 (kcal/kg)	Y_2 (%)	Y_3 (%)
1	0A	50	290	6907	51.50	1.37
2	+−	80	210	6433	72.57	1.28
3	a0	20	250	6250	70.25	1.31
4	++	80	290	6627	47.44	1.31
5	00	50	250	6880	55.24	1.36
6	−+	20	290	6715	40.96	1.33
7	0	50	250	6995	62.37	1.39
8	A0	80	250	6690	60.15	1.33
9	0a	50	210	6658	80.42	1.32
10	−−	20	210	6010	85.22	1.19

According to the experimental data, after press cake of camellia seeds are torrefied, the best calorific value (Y_1) appears in the seventh group of experimental conditions, the best energy density (Y_2) appears in the tenth group of experiments, and the best energy yield (Y_3) appears in the seventh groups of experiments. If the seventh group of operating conditions (50min and 250°C) is selected, Y_1, Y_2 and Y_3 have acceptable values.

Table 3. Experimental data of wood pellets.

NO.	Pattern	X_1 (min)	X_2 (°C)	Y_1 (kcal/kg)	Y_2 (%)	Y_3 (%)
1	−+	20	290	5863	42.40	1.41
2	0	40	250	5704	43.55	1.38
3	0A	40	290	5925	41.5	1.43
4	−−	20	210	4193	80.88	1.01
5	A0	60	250	5894	50.33	1.42
6	a0	20	250	5715	45.43	1.38
7	0a	40	210	4595	67.52	1.11
8	++	60	290	6338	41.23	1.53
9	0	40	250	5586	52.23	1.35
10	+−	60	210	4595	67.52	1.11

According to the experimental data, after wood pellets are torrefied, the best calorific value (Y_1) appears in the eighth group of experimental conditions, the best energy density (Y_2) appears in the fourth group of experiments, and the best energy yield (Y_3) appears in the eighth groups of experiments. If the eighth group of operating conditions (60 min and 290°C) is selected, Y_1, Y_2 and Y_3 have acceptable values.

Table 4. Experimental data of rice straw.

NO.	Pattern	X_1 (min)	X_2 (°C)	Y_1 (kcal/kg)	Y_2 (%)	Y_3 (%)
1	0a	40	210	4308	52.29	1.11
2	a0	20	250	4172	45.81	1.07
3	−+	20	290	4488	46.34	1.16
4	00	40	250	4415	50.13	1.14
5	++	60	290	4500	48.97	1.16
6	0A	40	290	4609	48.51	1.19
7	A0	60	250	4334	50.12	1.12
8	00	40	250	4372	47.48	1.13
9	−−	20	210	4203	50.50	1.08
10	+−	60	210	4259	60.15	1.10

According to the experimental data, after rice straw is torrefied, the best calorific value (Y_1) appears in the sixth group of experimental conditions, the best energy density (Y_2) appears in the tenth group of experiments, and the best energy yield (Y_3) appears in the sixth groups of experiments. If the eighth group of operating conditions (40min and 290°C) is selected, Y_1, Y_2 and Y_3 have acceptable values. Of course, these experimental data obtained by RSM design in Tables 1-4 can also be optimized by Analysis of Variances (ANOVA). Through the optimal discussion of the above response surface methodology, the optimal calorific value of pennisetum is at 276.17°C for a residence time of 73 min, and the optimal calorific value of press cake of camellia oil seeds is at 298°C for a residence time of 48.9 min. The properties of agricultural waste and the empty bunch of palm fruits are similar, so the palm was tested at a temperature of 298°C for a residence time, and the calorific value was found to be 5081 kcal/kg, which was about 31% higher than the initial calorific value of 3891 kcal/kg of untorrefied palm-waste RDF-5, indicating that low temperature carbonization is an effective thermal pretreatment method for biomass.

From the above results, it can be seen that temperature and residence time have a certain degree of influence on the calorific value, but the influence trends are not consistent in four materials. It can be found that the higher temperature, the higher calorific value, while the residence time has different effects. It can be known that the temperature has a great influence on the energy yield. Because the energy yield takes into account the weight comparison of the raw materials before and after carbonization. The lower the carbonization temperature, the smaller the weight change and the higher the energy yield, and the time has little effect. It can be found that for energy density, except pennisetum, time is a significant factor, however, temperature is a significant factor for the other biomasses, indicating that different biomasses have different effects on temperature and residence time. The higher the temperature, the higher the value of the energy density, and the highest energy density is found at a temperature of about 290 °C.

4 Conclusion

This experiment is to investigate the effects of four different materials on low-temperature carbonization (i.e. torrefaction) at different temperatures and different residence times. The raw materials are particles with a particle size of less than 350 μm, fed with a stable nitrogen flow, and the combustion furnace is an indirect electric heating tubular furnace for torrefaction. The test results include the structural characteristics of raw materials, optimal operating parameters, and weight structure changes before and after carbonization, and the following conclusions are obtained.

Among the four raw materials, wood chips, pennisetum and press cake of seeds, all have high volatile content and low ash content. The ash content of wood chips is the lowest at 2.98 wt%, and rice straw has the highest ash content among the four materials. If we want to use herbal biomass as a raw material for RDF-5, the boiler sintering problem and the cost of the ash removal system need to be considered. In calorific value analysis of raw materials, press cake of oil seeds has a higher calorific value than other substances, and its carbon content is also the highest in elemental analysis, indicating that carbon content is the main source of heat generation. The nitrogen and sulphur contents of the four materials are not too high, which can avoid the generation of sulphur oxides (SO_X) and nitrogen oxides (NO_X) during the combustion process, indicating that the four materials are suitable as fuels of RDF-5.

Comparing the highest calorific value of the four materials after low-temperature carbonization with the initial calorific value, pennisetum increased by about 42.5%, wood chips increased by 52.7%, press cake of oil seeds increased by 38.7%, and rice straw increased by as little as 18.6% , because the carbonization is mainly the decomposition of cellulose and hemicellulose, it can also be found in the approximate analysis that the volatile content of rice straw is low, so the calorific value is low. Temperature and residence time have a certain degree of influence on calorific value, but the influence trends are not consistent. It can be found that the higher the temperature, the higher the calorific value. The calorific value of the four materials can reach the highest at 290 °C, while the effect of residence time is less consistent. Except pennisetum, for energy density which is a significant factor for residence time, temperature is a significant factor for the rest of the biomasses, indicating that different biomasses have different effects on temperature and residence time, but it can be found that the higher the temperature the higher the value of the energy density and the highest energy density exists at a temperature of about 290 °C.

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