

## Co-pyrolysis of rubberwood sawdust (RWS) and polypropylene (PP) in a fixed bed pyrolyzer

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### ABSTRACT

Co-pyrolysis of rubberwood sawdust (RWS) waste and polypropylene (PP) was carried out at different temperatures (450,500,550, and 600°C) with biomass to plastics ratio 1:1 by using fixed bed drop-type pyrolyzer. The yield of pyrolysis oil has an increasing trend as the temperature increased from 450°C to 550°C. However, the pyrolysis oil yield dropped at a temperature of 600°C. Co-pyrolysis of RWS and PP generated maximum pyrolysis oil with 36.47 wt.% at 550°C. The result is compared with the pyrolysis of RWS only without plastics, with the same feedstock, and the maximum pyrolysis oil yield obtained was 33.3 wt.%. The water content in pyrolysis oil of co-pyrolysis RWS with PP is lower than RWS only with 54.2 wt.% and 62 wt.% respectively. Hydrocarbons, acyclic olefin, alkyl, and aromatic groups are the major compound in the pyrolysis oil from the co-pyrolysis process. Carbon monoxide (52.2 vol.%) and carbon dioxide (38.2 vol.%) are the major gas components.

**Keywords:** Co-pyrolysis; Rubberwood sawdust residue; Polypropylene; fixed-bed drop type pyrolyzer

### INTRODUCTION

As the world is reaching peak oil usage, the depletion of fossil fuel is imminent. Alternative energy source should be sought to prepare us for the uncharted future where fossil fuel is scarce. Biomass is considered a carbon-neutral energy source that is not hazardous to the environment since it does not increase CO<sub>2</sub> concentration in the atmosphere during the energy conversion process. It also has high volatile matter content and low contents of nitrogen, sulphur, and ash that can contribute pollution to the environment when burned [1].

Biomass can be cultivated exclusively for energy production such as corn and sugarcane, however, the problem arises from the usage of land that can be otherwise be used to grow food, thus creating the debate of food vs fuel [2]. To avoid this problem, the biomass source from agricultural residue is preferred. Cash crop such as oil palm and rubber tree is widely cultivated for the production of palm oil and latex, and this industry will produce a significant amount of waste as a potential biomass source for energy production [3-4].

Rubber tree has been widely cultivated in ASEAN countries such as Thailand, Indonesia, Vietnam, and Malaysia, with the amount of natural rubber production are 4 million tonnes, 1 million tonnes and 600 thousand tonnes respectively [5]. After 25 years of the latex yielding, the rubber trees are cut down and the wood could be utilized for many downstream processed such as sawmilling, furniture, panel products, builders carpentry and joinery. In Malaysia alone, the production of rubberwood biomass waste is recorded at  $1.5 \times 10^6 \text{ m}^3$  in volumes, with an estimation energy content of 76 million GJ [3].

Pyrolysis is one of the potential processes to elevate rubberwood residue as valuable added fuels. Pyrolysis process is defined as the direct thermal decomposition of the organic materials in the absence of oxygen. Pyrolysis of rubber wood residue waste was studied by Mazlan et al [6]. They found out as the temperature increased the yield of pyrolysis oil and NCG increased. Maximum pyrolysis oil at 33.0 wt% was generated at temperature  $550^\circ\text{C}$  and when the temperature increased, continues volatilization reaction takes place caused the yield of biochar decreased. However, in general, pyrolysis oil has poor properties, such as low calorific value, high oxygen content [7], high water content, corrosiveness and instability [8], [9], which resist its direct application as a transportation fuel and thus, need upgrading before application. Co-pyrolysis process has been suggested by researchers because of its potential in producing high-grade pyrolysis oil [10]–[12].

The presence of plastics as co-feeds act as good hydrogen source during pyrolysis which leads to a decrease in oxygen content in pyrolysis oil. As plastics have significantly lower water content than biomass and do not absorb moisture [13], a costly drying process is not needed. Aside from upgrading the pyrolysis oil, adding plastics can also solve the problem of plastic wastes manage. Plastic waste management has been a problem, as domestic plastic usage is on the rise. In 2015, 6300 Mt of plastic has been produced, and plastics waste in landfill is expected to reach 12,000 Mt on 2050 if this trend continues[14]. Polyethylene and polypropylene are the two of the most produced types of non-fiber plastics, with the share as large as 36% and 21% respectively [14]. Exploring the use of rubberwood residue mixed with plastics is therefore beneficial as both feedstocks are abundantly available. However, no specific research has been done for this combination of feedstocks.

In this work, rubberwood residue was co-pyrolyzed with polypropylene (PP) at ratio 1:1 using a fixed-bed drop type pyrolyzer. To identify the upgrading effect of the presence of PP in the blend of feedstock, the solid, liquid and gas production is compared with the pyrolysis of RWS only.

## **EXPERIMENTAL SET UP**

### **Feedstock**

The biomass used in this work is the rubberwood residue (RWS) which is collected from a sawmill in Perak, Malaysia. The raw material was in the form of small particles and sieved to a size range of 0.15-0.50 mm as it favors higher pyrolysis reaction [15]. Prior to the experiment, the RWS was dried in a drying oven for 24 h at  $105^\circ\text{C}$  to diminish moisture. Polypropylene (PP) was collected from a food container waste and was cut into sizes of 0.3 mm x 0.3 mm. The characteristics of the feedstocks are shown in Table 1.

Table 1. Characteristics of feedstocks

Characteristics	RWS	PP
Proximate analysis (wt. %)		
Moisture	4.3	-
Ash	1.8	-
Volatile content	79.8	99.2
Fixed carbon	14.1	0.8
Elemental analysis (wt. %)		
C	44.2	87.8
H	7.2	10.6
N	0.3	0.2
O	48.3	1.4
Calorific value (kJ/g)	18.9	46.3
Hemicellulose (wt.%, Dry) [16]	34.60	-
Cellulose (wt.%, Dry) [16]	18.30	-
Lignin (wt.%, Dry) [16]	25.50	-

### Experimental setup

Experiments were carried out using fixed-bed, drop type pyrolyzer as shown in Figure 1. The reactor was connected to nitrogen gas lines and vacuum to create an inert condition. Inert conditions were achieved by purging nitrogen three times, to ensure the total removal of oxygen or air in the system. The pyrolyzer was designed with two ball valves at the biomass holder zone. The drop-type pyrolyzer is made of a stainless steel cylindrical reactor with a size of 155 mm height and 53 mm internal diameter. The reactor was placed inside the electrical heater and was insulated to minimize heat loss during the experiment. A K-type thermocouple was embedded inside the reactor to measure the pyrolysis temperature.

Two condensers were connected to the reactor to collect the liquid product. Both condensers were immersed in the cool water bath. The sampling gas bag was connected to the outlet of the vapour trap to accumulate the non-condensable gases (NCG).

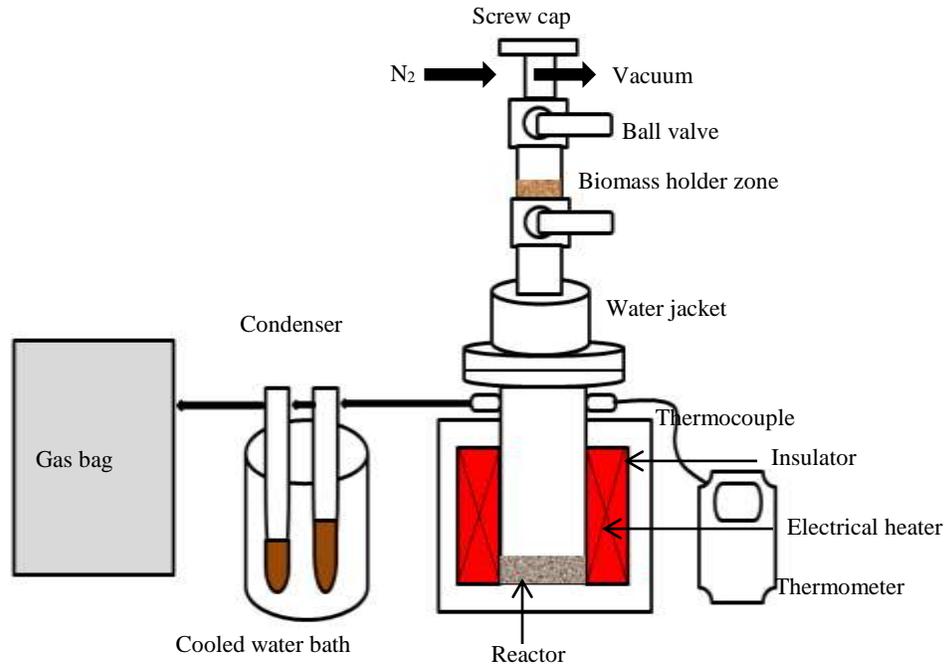


Figure 1. Fixed-bed drop type pyrolyzer schematic diagram

**Experimental procedure**

The co-pyrolysis experiment was accomplished with 1:1 RWS to PP mass ratio at different temperatures. Each experiment was conducted three times to ensure consistency. After the reactor was set in an inert condition by purging Nitrogen gas, the reactor was heated to the desired temperature, and then the feedstocks were dropped down by gravity by opening the ball valve. The experiment was performed using 15 g of feedstocks at the yield of pyrolysis oil, biochar, and NCG products were collected. The duration of the experiment was held for approximately 15 minutes until no production of gas fumes was observed. The flowchart for the summary of experimental procedure is shown in Figure 2.

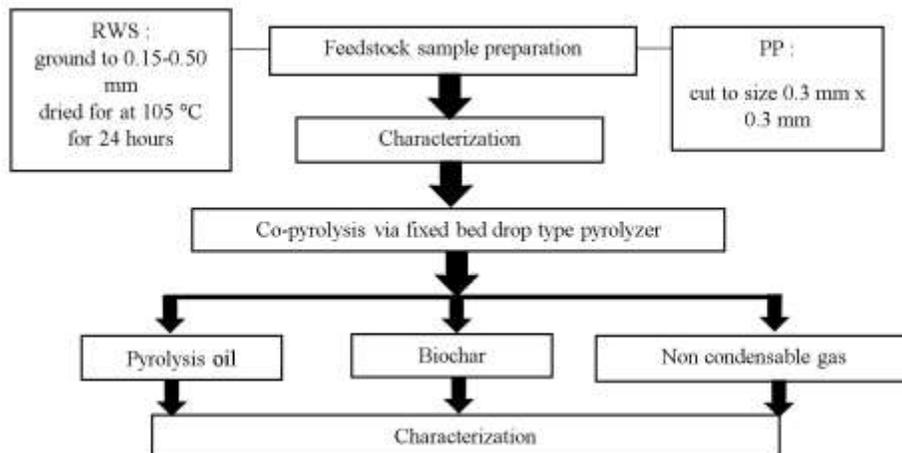


Figure 2. Experimental procedure flow chart

### Characterization of feedstocks and co-pyrolysis products

To study the effect of feedstock on the quantities of the quantities of co-pyrolysis product, the analysis of oil products from the maximum pyrolysis temperature was executed. The water content of pyrolysis oil was evaluated by a volumetric Karl Fisher titrator (Mettler Toledo, V30). The elemental analysis was accomplished using a Perkin Elmer 2400 CHN Analyzer. The value of Hemi-cellulose, cellulose and lignin of RWS is referred to published literature [16]. The calorific value was computed by an IKA WERKE (2000) bomb calorimeter to calculate the energy contained in pyrolysis oil and biochar. Gas Chromatograph/ Thermal Conductivity Detector (GC-TCD) (Shimadzu GC-8A) was used to define the composition of non-condensable gases, with a Davison Grade 12 Silica Gel packed column to detect CO, CH<sub>4</sub>, O<sub>2</sub>, and H<sub>2</sub>. The organic compound of the pyrolysis oil was detected by using a Gas Chromatograph Mass Spectrometry (GC-MS). The GC-MS analysis was carried out with an Agilent Technology model 7890A series. A BPX-5 capillary column was selected with 0.25mm, 30mm and 0.25 $\mu$ m of its internal diameter, length and film thickness correspondingly. Thermogravimetric analysis (TGA) was done on the RWS and PP feedstock respectively using the TA Instruments model QA50 with 60 mL/min of N<sub>2</sub> flowrate at a heating rate of 10 °C/min. Figure 3 and figure 4 shows the thermogravimetric curves for the RWS and PP plastic respectively. Around 220 °C, the RWS started to break down, whereas PP starts to decompose approximately at 400°C as seen in Figure 2 and Figure 3 respectively.

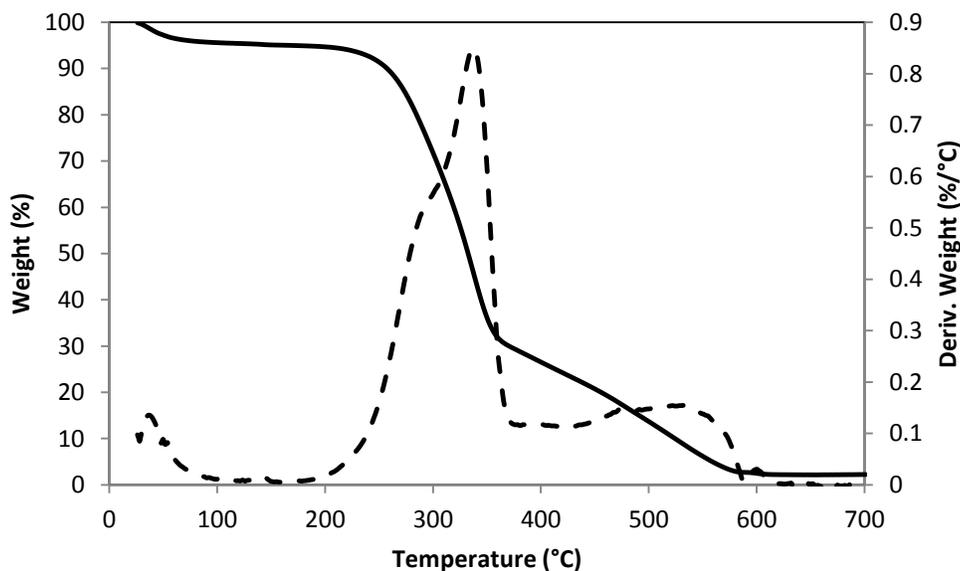


Figure 3. TGA of RWS feedstock

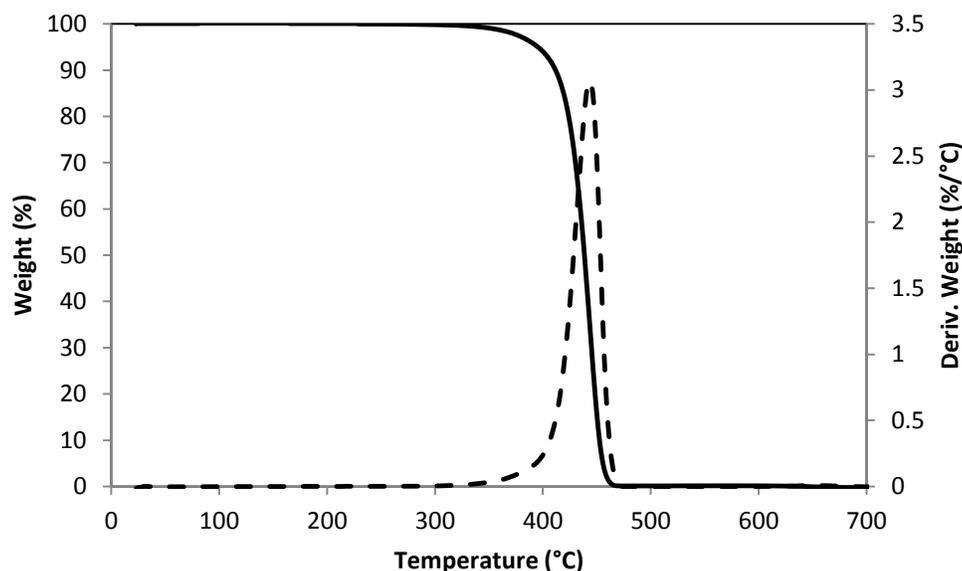


Figure 4. TGA of PP feedstock

## RESULTS AND DISCUSSION

### Effect of temperature on co-pyrolysis products

To study the effect of temperature on the co-pyrolysis products, the experiment was held at 450 °C, 500 °C, 550 °C, and 600 °C. These temperatures were selected as from the TGA results, the pyrolysis of RWS and PP starts at 220 °C and 400 °C respectively. In order for both of the feedstock to decompose, temperature above 400 °C was used. The yield of resulting products from co-pyrolysis of RWS and PP were discussed for examining the effect of pyrolysis temperature on 1:1 feedstock ratio. The co-pyrolytic yield from RWS and PP produced pyrolysis oil, biochar, and non-condensable gases.

As shown in Figure 5, the yield of pyrolysis oil has an inclined trend as the temperature increased from 450°C to 550°C. This can be explained as the higher temperature will cause greater primary decomposition of the feedstock and secondary decomposition of char. This also explained the decrease of char with increasing temperature. Maximum pyrolysis oil yield is achieved at 550°C with 36.47 wt%. However, the pyrolysis oil yield abruptly dropped at a temperature of 600°C, together with a further decrease of char, producing more non-condensable gas. The very high temperature will promote further cracking of condensable vapour to non-condensable gas [17], explaining the decrease of pyrolysis oil yield.

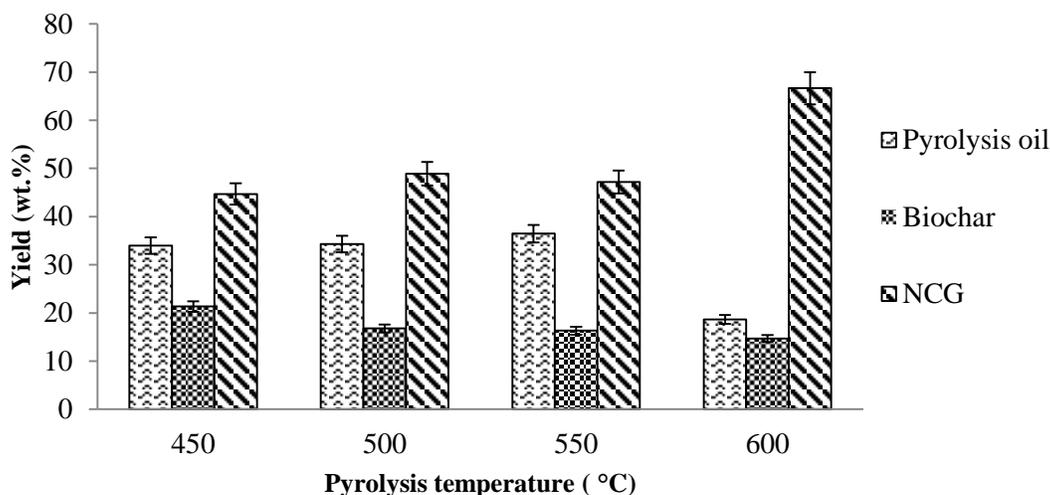


Figure 5. Product yield for the co-pyrolysis of RWS with PP

However, the comparison between pyrolysis of RWS alone was done with the same weight to compare the product yields with co-pyrolysis product yields. Pyrolysis of RWS only in Figure 6 shows the pyrolysis oil is lower compared to co-pyrolysis process. The Maximum pyrolysis oil of pyrolysis RWS only at temperature 550°C with 33.3 wt.% compared to co-pyrolysis with PP at 36.47 wt.%. As PP contains more volatile matter, adding PP to the feedstock will yield more liquid and gaseous product with less char yield.

The same trend was observed for the effect of temperature on the pyrolysis of RWS only and co-pyrolysis oil RWS with PP. As the temperature increased, the pyrolysis oil and NCG increased while biochar decreased, until a certain point in which when higher temperature is used, the pyrolysis oil yield will decrease.

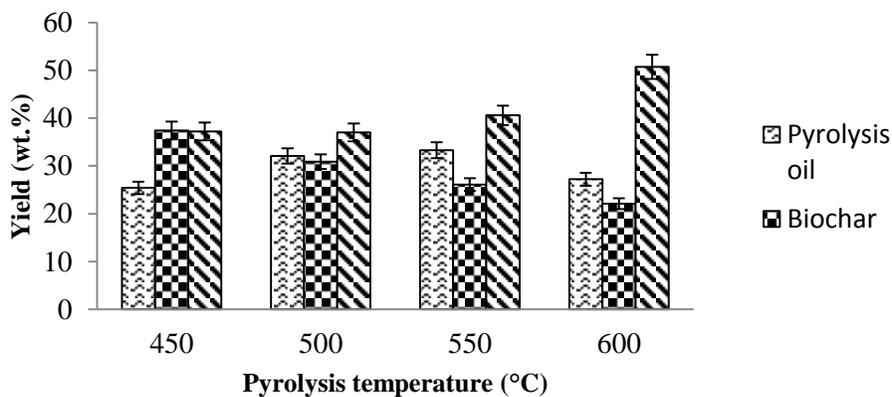


Figure 6. Product yield for pyrolysis of RWS only

### Characteristics of products

The pyrolysis oil collected from pyrolysis process were red-brown coloured and had an acrid odour. Table 2 summarized the characteristics and properties of pyrolysis RWS only and co-pyrolysis of RWS with PP products.

Product yields from the maximum pyrolysis temperature at 550°C for the pyrolysis of RWS only and RWS and PP were used to products analysis.

Table 2. Characteristics and properties analysis of products yields

Characteristics	RWS only		Co-pyrolysis with PP	RWS
	Pyrolysis oil	Bio-char	Pyrolysis oil	Bio-char
Proximate analysis (wt.%)				
Moisture		2.7		2.6
Ash	-	4.6	-	5.2
Volatile	-	51.2	-	54.1
Fixed carbon	-	41.5	-	38.1
Elemental analysis (wt.%)				
C	19.7	74.5	43.1	78.2
H	10.2	2.5	13.9	1.5
N	0.1	0.4	0.1	0.5
O	70.0	22.6	42.9	19.8
Water content (wt.%)	62.0	-	54.3	-
Calorific value (kJ/g)	7.2	27.6	14.8	29.4

It can be observed that the water content in pyrolysis oil of co-pyrolysis RWS with PP is lower than RWS only with 54.2 wt.% and 62 wt.% respectively. The presence of PP in the co-pyrolysis process with RWS can balance the C, H, and O in the feedstock, with cogent effect on the properties of degradation products [18]. To control the water content of pyrolysis oil, some researcher concluded that drying the feedstock (approaching zero) show to decrease water content [19]. High water content in pyrolysis oil is an unsuitable component because it can be destructive for the ignition performance. High water content also affects the calorific value of the pyrolysis oil and might reduce it potentially as energy [20].

In addition, hydrogen is one of the important components with a point to increase the calorific value because it has the highest heating value [21]. Rubberwood is categorized as a hardwood which has higher lignin content and might have relatively higher biochar yields [22]. The biochar yields of co-pyrolysis have high carbon content and low oxygen content, thus biochar may be used as carbon fibers, soil fertilizer, chemicals, activated carbon, etc. [20], [23].

Variation in organic yields is due to differences in the physical and chemical composition of feedstock. The liquid fractions were composed by a large number of compounds and most of them in a low concentration below 0.1 (v/v). Over hundreds of organic compounds was found in the pyrolysis oil but only 0.1 (v/v) and above were listed in Table 3. Hydrocarbons, diolefin, aromatic and carboxylic acid groups are the major compound in the pyrolysis oil from the co-pyrolysis process. 2,4-dimethyl-1-heptene was found to be the highest with 8.05 (v/v) in pyrolysis oil. Alkenes such as hexane, pentene and octane can also be detected in the oil. This result is an agreement with result obtained by Sophonrat et al, 2017, when cellulose is co-pyrolyzed with PP plastic [24]. Comparing with

pyrolysis of RWS only [6], mixing PP to RWS increased longer chain hydrocarbons in the pyrolysis oil. Similar result is obtained by Ballice et al 2002 when co-pyrolysis experiment is done with lignite and PP [25].

Table 3. Organic compounds in pyrolysis oil from co-pyrolysis of RWS with PP.

Compound	%Area	Compound	%Area
<b>Alkanes</b>		<b>Diolenes</b>	
Cyclobutane (C <sub>4</sub> H <sub>8</sub> )	0.55	2,4-Hexadiene (C <sub>6</sub> H <sub>10</sub> )	0.22
Cyclopropane (C <sub>3</sub> H <sub>6</sub> )	0.58	1,3-Pentadiene (C <sub>5</sub> H <sub>8</sub> )	0.91
Eicosene ([CH <sub>2</sub> ] <sub>18</sub> -CH <sub>3</sub> )	0.23	1,5-Hexadiene (C <sub>6</sub> H <sub>10</sub> )	0.71
Cyclo-octacosane (C <sub>28</sub> H <sub>56</sub> )	0.21	1,4-cyclohexadiene (C <sub>6</sub> H <sub>8</sub> )	0.24
Cyclohexane (C <sub>6</sub> H <sub>12</sub> )	1.23	1,4-Hexadiene (C <sub>6</sub> H <sub>10</sub> )	0.42
2,3-Dimethyl-3-heptane (C <sub>8</sub> H <sub>16</sub> )	1.65	1,5-Heptadiene (C <sub>7</sub> H <sub>12</sub> )	0.37
Octane (C <sub>8</sub> H <sub>18</sub> )	0.41	2-Methyl-1,5-Heptadiene (C <sub>7</sub> H <sub>12</sub> )	0.20
Heptane (C <sub>7</sub> H <sub>16</sub> )	1.07	1,3-Hexadiene (C <sub>6</sub> H <sub>10</sub> )	0.15
<b>Alkenes</b>		<b>1,4-Hexadiene (C<sub>6</sub>H<sub>10</sub>)</b>	
2-Heptene (C <sub>7</sub> H <sub>14</sub> )	0.28	1,7-Nonadiene (C <sub>9</sub> H <sub>16</sub> )	0.17
1,3,5-Hexatriene (C <sub>6</sub> H <sub>8</sub> )	0.95	1,12-Tridecadiene (C <sub>13</sub> H <sub>24</sub> )	0.19
2,4-Dimethyl-1-heptene (C <sub>9</sub> H <sub>18</sub> )	8.05	<b>Esters</b>	
1,3-Dimethylcyclohexene (C <sub>8</sub> H <sub>14</sub> )	0.20	Octacosyl Trifluoroacetate (C <sub>30</sub> H <sub>57</sub> F <sub>3</sub> O <sub>2</sub> )	0.82
Heptene (C <sub>7</sub> H <sub>14</sub> )	0.12	<b>Carboxylic Acids</b>	
2-Octene (C <sub>8</sub> H <sub>16</sub> )	1.30	Oxalic acid (C <sub>2</sub> H <sub>2</sub> O <sub>4</sub> )	0.22
2-Undecene (C <sub>11</sub> H <sub>22</sub> )	0.27	Fumaric acid (C <sub>4</sub> H <sub>4</sub> O <sub>4</sub> )	0.16
Cyclodecene (C <sub>10</sub> H <sub>20</sub> )	0.15	Cyclopropanecarboxylic acid (C <sub>4</sub> H <sub>6</sub> O <sub>2</sub> )	0.32
Dodecene (C <sub>10</sub> H <sub>21</sub> CH=2)	0.17	2-Butenoic Acid	0.94
Cyclododecane ((CH <sub>2</sub> ) <sub>12</sub> )	0.18	Acetic acid (CH <sub>3</sub> COOH)	0.22
1-Heptadecene (C <sub>17</sub> H <sub>34</sub> )	0.20	<b>Aromatics</b>	
3-Octadecene (C <sub>18</sub> H <sub>36</sub> )	0.24	Ethylbenzene (C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub> CH <sub>3</sub> )	0.83
Pentene (C <sub>5</sub> H <sub>10</sub> )	1.13	2-Methylindene (C <sub>10</sub> H <sub>10</sub> )	0.28
Cyclohexene (C <sub>6</sub> H <sub>10</sub> )	1.37	Benzene (C <sub>6</sub> H <sub>6</sub> )	4.12
Toluene (C <sub>7</sub> H <sub>8</sub> )	4.29	<b>Phenols</b>	
Hexene (C <sub>6</sub> H <sub>12</sub> )	1.55	Phenol (C <sub>6</sub> H <sub>5</sub> OH)	0.21
2-pentene (C <sub>5</sub> H <sub>10</sub> )	1.91	<b>Phenyls</b>	
2-octene (C <sub>8</sub> H <sub>16</sub> )	1.30	3-Aminopyrazole (C <sub>3</sub> H <sub>5</sub> N <sub>3</sub> )	0.87
<b>Benzyl Alcohols</b>		<b>Hydroxyls</b>	
Benzenemethanol (C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub> OH)	0.26	Cyclohexanol (HOCH(CH <sub>2</sub> ) <sub>5</sub> )	0.30
<b>Ketones</b>		Cyclopropylmethanol (C <sub>4</sub> H <sub>8</sub> O)	0.70
Cyclopentanone ((CH <sub>2</sub> ) <sub>4</sub> CO)	0.30	<b>Polycyclic Aromatics</b>	
Ethanone (C <sub>10</sub> H <sub>16</sub> O)	0.70	Naphthalene (C <sub>10</sub> H <sub>8</sub> )	0.19
<b>Benzoate</b>		<b>Silane</b>	
Benzoic acid (C <sub>7</sub> H <sub>6</sub> O <sub>2</sub> )	0.48	Triallylsilane (C <sub>9</sub> H <sub>15</sub> Si)	0.62
<b>Acyclic olefin</b>			
4-methyl-2-heptene (C <sub>8</sub> H <sub>16</sub> )	1.90		

Non-condensable gases (NCG) were collected in the gas bag and the composition analysis is often disregarded by many researchers. NCG has brownish vapour colour and has an acrid odor. Figure 7 shows the gas composition analysis of NCG. In this study, carbon monoxide (52.2 vol.%) and carbon dioxide (38.2 vol.%) were the major gas components.

■ Unknown gas ■ Carbon dioxide ■ Carbon monoxide ■ Methane ■ Hydrogen

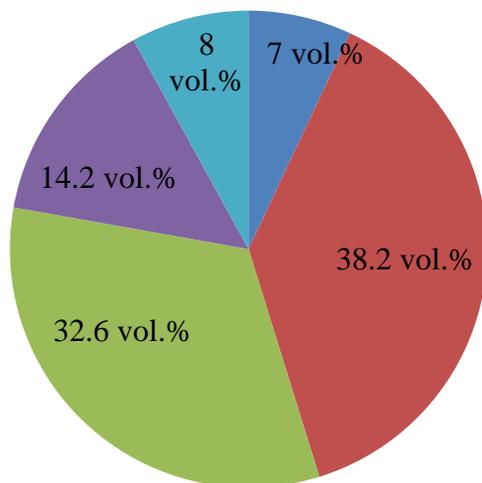


Figure 7. Non-condensable gas (NCG) composition

## CONCLUSION

In this work, the effect of co-pyrolysis of RWS with PP at different temperatures with 1:1 ratio was investigated. The products yields of pyrolysis oil, biochar, non-condensable gases and their composition were discussed. The yield of pyrolysis oil has an inclined trend as the temperature increased from 450°C to 550°C because of the continuous pyrolysis conversion at the higher temperature. However, the pyrolysis oil abruptly dropped at temperature 600°C, assuming and producing more gases. Co-pyrolysis of RWS and PP generated maximum pyrolysis oil with 36.47 wt.% at 500°C. However, the comparison between pyrolysis of RWS alone was done with the same weight to compare the product yields with co-pyrolysis product yields and maximum pyrolysis oil yield obtained was 33.3 wt.%. The water content in pyrolysis oil of co-pyrolysis RWS with PP is lower than RWS only with 54.2 wt.% and 62 wt.% respectively. Hydrocarbons, acyclic olefin, alkyl and aromatic groups are the major compound in the pyrolysis oil from the co-pyrolysis process. Carbon monoxide (52.2 vol.%) and carbon dioxide (38.2 vol.%) were the major gas components.

It can be concluded that mixing RWS with PP as the feedstock has many advantages. Apart from higher pyrolysis oil yield, the quality of the oil was also improved with higher calorific value and lower water content. For future research, investigation of co-pyrolysis of RWS and PP in a larger scale continuous fluidized-bed pyrolyzer is important to fully understand this process in a larger scale.

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