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CHARACTERIZATION, CALCULATION OF CALORIFIC VALUES, AND BIO-OIL PRODUCTION VIA THERMOCHEMICAL PROCESSES OF MUNICIPAL SOLID WASTE IN PERLIS, MALAYSIA

(Pencirian, Pengiraan Nilai – Nilai Kalorifik dan Pengeluaran Bio-Minyak Melalui Proses Termokimia Sisa Pepejal Perbandaran di Perlis, Malaysia)

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Abstract

Municipal solid wastes (MSW) collected from Padang Siding Landfill, Perlis were segregated and characterized in the laboratory. The main components of MSW found are paper, plastic, glass and large proportion of organic waste. Moisture content was measured for all the components. Paper and yard wastes recorded the highest percentage of 26.7% and 28.8%, respectively. Thermogravimetric analysis (TGA) shows that the thermal degradation of MSW samples ranged from 200 °C to 800 °C, indicating fast decomposition occurred at 370 °C, 430 °C and 700 °C. Traditional method shows a lower error compared to the physical method in calculating the calorific values (CV) for MSW. Bio-oil production via fast pyrolysis route gave higher liquid yield of ~35% with high oxygen content of ~49 – 53%. Heavy oil recovered from hydrous pyrolysis experiments gave low yield of ~7 – 13%, however with low oxygen content of ~11 – 18%.

Keywords: Municipal solid waste, thermogravimetric analysis, calorific value, pyrolysis, hydrous pyrolysis

Abstrak

Sisa pepejal perbandaran (SPP) dikumpulkan dari tapak pelupusan Padang Siding, Perlis. Bahan buangan ini diasingkan dan dikategorikan di dalam makmal. Komponen utama MSW yang ditemui adalah kertas, plastik, kaca dan sebahagian besar sisa organik. Kandungan kelembapan diukur untuk semua komponen di mana kertas dan sisa dari laman telah mencatatkan peratus tertinggi sebanyak 26.7% dan 28.8% masing-masing. Analisis termogravimetrik menunjukkan degradasi terma sampel MSW adalah antara 200 °C hingga 800 °C, menunjukkan penguraian cepat berlaku pada 370 °C, 430 °C dan 700 °C. Kaedah tradisional menunjukkan ralat yang lebih rendah berbanding dengan kaedah fizikal dalam mengira nilai kalorifik untuk SPP. Pengeluaran bio-minyak melalui laluan pirolisis pantas memberikan hasil cecair yang lebih tinggi ~35% dengan kandungan oksigen yang tinggi ~ 49 – 53%. Minyak berat diperoleh daripada eksperimen pirolisis berair memberikan hasil yang rendah ~ 7 – 13%, walau bagaimanapun dengan kandungan oksigen yang rendah ~ 11 – 18%.

Kata kunci: Sisa pepejal perbandaran, analisa termogravimetrik, nilai kalorifik, pirolisis, pirolisis berair

Introduction

Municipal solid waste (MSW), also known as urban solid waste, is a type of waste that predominantly comes from household waste (domestic waste) with a small portion of commercial waste collected by municipality of a particular area. Generally, the solid waste is defined as the household's refusal and non-hazardous solid waste from industrial, commercial and institutional establishments, such as hospitals, market and yard wastes, and street sweepings [1]. MSW is a heterogeneous material and its physical composition is dependent on socio-economic level and climatic conditions [2].

Perlis Indera Kayangan is the smallest state in Malaysia which is located at the northern part of the Peninsular Malaysia. Its population is 227,025 as of 2010. The increase in population and industrial activities in Perlis has resulted in serious environmental problems including air quality deterioration, water and land pollutions. The amount of solid wastes generated in Perlis is approximately 221 tonnes/day [3]. The most popular disposal practice for MSW is landfilling. Landfilling is a universal solution that provides ultimate waste disposal and can be implemented easily. However, the setback of this approach is that it requires a large piece of land and does not employ the concepts of reduction and conversion of MSW. The MSW disposal in Perlis is located at Padang Siding landfill with an area approximately 50.876 hectare. Landfills face the problems of overfilling, overflowing of leachates leading to pollution of water resources, and uncontrolled dust emissions adversely affecting the human environment [4].

In order to justify the feasibility of energy recovery for the use of renewable energy, it is very important to determine the calorific value (CV) of MSW. Bomb calorimeter can be used for calorimetric measurement in which combustion is conducted under conditions of constant volume. Other important methods are physical composition and traditional methods where the energy content can be calculated using physical and proximate analysis, respectively.

One option of utilizing the MSW is energy recovery through various processes, such as combustion, pyrolysis, liquefaction and refuse-derived fuel [5]. Several researchers have pyrolyzed biomass residue to produce bio-oil under certain conditions [6 – 8]. Bio-oil is dark brown liquid which has a high heating value that can be used as fuels in boiler, diesel engines for power generation, or upgraded to produce fuels and bulk chemicals. However, direct usage of bio-oil as conventional fuels may present some difficulties due to high viscosity, poor heating value, corrosiveness and instability. The comparison of pyrolysis and hydrous pyrolysis of MSW has not been done in earlier studies and this study attempts to fill in the gap. The comparison is beneficial for engineers and policy makers to design an alternative solution for MSW.

A large portion of the waste has significant energy potential which can be utilized. The energy recovered can also reduce emissions that would otherwise be produced by other energy systems, such as fossil fuel thermal power plants [9]. An important criterion for the success of waste management plan is the accurate data on the quality and quantity of the waste generation. With this data, proper management strategies can be planned and put into actions. The data can also be used to predict the future trends of the quantity and quality of the MSW. This allows the authorities to take anticipating measures to manage MSW generated properly [10].

In order to justify the feasibility of energy recovery for the use of renewable energy, it is very important to determine the calorific value (CV) of MSW. Bomb calorimeter can be used for calorimetric measurement in which combustion is conducted under the condition of constant volume. Other important methods are physical composition and traditional methods where the energy content can be calculated using physical and proximate analysis, respectively.

In Perlis, there is less data available on the potential of MSW to be used as energy sources. To fill in the gap, this study was conducted to segregate and characterize the main components of MSW in the state. The energy contents of MSW generated were calculated. The production of bio-oil was performed via thermochemical conversion processes to investigate its potential to be used as a renewable energy.

Materials and Methods

Sample preparation

The solid waste samples were collected at Padang Siding landfill in Arau, Perlis. The sampling was carried out randomly at 5 different locations within the 0.12 km^2 landfill area of 0.6 m depth. These samples were brought back to the laboratory for analysis. The manual segregation process was conducted to calculate the MSW's components. After the completion of segregation, all the samples were mixed, washed (with water) and oven dried at 105 ^{o}C for 12 hours to eliminate external moisture. The samples (except glass) were shredded and sieved to obtain uniform size less than 1.5 mm for further experimental works. This sample was used for TGA analysis and pyrolysis experiments.

Moisture-content determination

The collected samples were oven-dried at 105 ^{o}C for 5 hours to measure moisture-content. All the moisture content values used in this research were based on the wet basis, unless otherwise stated. The percentage of moisture content was calculated as follow:

$$Moisture\ (\%) = \left[\frac{Wet\ weight - dry\ weight}{Wet\ weight}\right] \text{ x } 100\% \tag{1}$$

Thermogravimetric analysis

The thermogravimetric analysis was carried out by using a TGA/DSC 1 Mettler Toledo. The weight loss and the rate of weight loss were recorded continuously under dynamic conditions, as functions of time or temperature, in the range of 25 to 900 ^{o}C. Purified nitrogen gas was used for the pyrolysis with a flow rate of 150 ml/min. The heating rate of 20 ^{o}C/min was selected for this test. The temperature was increased to the setting value of 900 ^{o}C at the pre-selected heating rate and it was kept constant at this value until steady conditions were obtained.

Proximate analysis

Proximate analysis was performed according to ASTM D2974 [11] using the thermogravimetric analyzer (TGA/DSC 1, Mettler Toledo) for determination of moisture, volatile matter, fixed carbon and ash in MSW. The experiment was performed under inert nitrogen gas and purified air with a constant flow rate of 150 ml/min and the heating rate of 20 ^{o}C. The samples were weighed directly into the alumina crucible and the temperature was kept at isothermal for 0.5 minutes until a steady condition was obtained before ramping to the desired temperature [12]. The experiments were replicated at least twice to obtain reproducibility.

Calorific value (CV)

The calorific values (CVs) of the samples were determined by using a bomb calorimeter (Model: C2000 basic, IKA co, Germany). The calculation was also made according to the model developed by Khan and Abu Ghrarah [13] as reported by Abu-Qudais and Abu-Qdais [5].

$$E = 23\ [F + 3.6\ (PA)] + 160\ (PL) \tag{2}$$

where E is the energy content of MSW (Btu/lb), PL is the percentage of plastic by weight, F is the percentage of food waste by weight and PA is the percentage of paper waste by weight.

For comparison, a traditional model reported by Abu-Qudais and Abu Qdais [5] had also been employed as follows:

$$E\ (\text{Kcal/kg}) = 45B - 6W \tag{3}$$

where B is the combustible volatile matter in MSW (%) and W is the water percentage weight on dry basis (%).

CHNO analyses

The elemental analyses of the oils were determined using Thermo Electron FlashEA 1112 elemental analyser. Vanadium pentoxide (2-3 mg) was used to determine the Sulphur content and the amount of Sulphur found was too small and negligible for the calculation of this research.

Pyrolysis experimental procedure

Fast pyrolysis was conducted in a fixed-bed reactor as in our previous study [7]. The experimental device consisted of a tube reactor with a volume of 250 cm^3, equipped with a K-type thermocouple , an electric heater, a nitrogen cylinder, a cold trap, water bath and product effluent. The condensation temperature was set at 0 ^{o}C for all experiments. The amounts of sample used in all experiments were 5 mg per run. The bio-oil yield was collected from oil collector and weighed to calculate the percentage. For all experiments, whenever the pyrolysis temperature was reached, it was immediately cooled down to 100 ^{o}C within 4 minutes. Experiments were repeated 5 times to ensure the reproducibility of the results.

Nitrogen flow rate and condensation temperature were kept constant at 100 cm^3/min and 0 ^{o}C, respectively. The pyrolysis experiments were conducted using fixed bed pyrolyser at pyrolysis temperature of 470 ^{o}C, particle size of 0.60 mm, holding time at 1 minute. Experiments were repeated 5 times to ensure the reproducibility of the results.

Hydrous experimental procedure

Hydrous experiments were conducted using a Parr 4740 series stainless steel (75 ml cylindrical) pressure vessel at pre-set temperatures (380, 420 and 480 ^{o}C) for 1 hours. After sealing the vessel and attaching the pressure gauge, it was purged 10 times with nitrogen gas to remove the air, and then 2 bar of nitrogen gas was introduced to provide an inert atmosphere. The reactor was heated in a fluidised sand bath. Temperature was monitored by an additional K-type thermocouple which was connected to a computer and recorded every 10 s. The reacted product washing procedure had been reported in the previous study [14]. Hydrous pyrolysis was also conducted at 380, 420 and 480 ^{o}C with sample to water mass ratio of 1:6, using 75 ml reactor for 1 hour. The pressures recorded for these experiments were in the range of 280 – 300 bar.

Results and Discussion

Waste segregation and moisture content

Table 1 shows the average compositions after the segregation process. Plastic and glass were the highest proportion in the samples collected with approximately 46%. Similarly, papers contributed approximately 21%, ranked as the third largest component. The other smallest components were polystyrene, yard wastes and wood with 1.4%, 3.5% and 8.0%, respectively. Food waste consists of approximately 12%, as such, it can be inferred that the management of organic waste has not been properly done.

Table 1. Characterization of MSW from Padang Siding landfill

Sample	Food Waste (%)	Paper (%)	Plastic (%)	Polystyrene (%)	Rubber (%)	Yard waste (%)	Glass (%)	Wood (%)
1	37.80	1.50	13.90	1.20	24.70	6.40	0.00	14.50
2	2.80	4.00	15.40	2.20	11.30	7.80	46.20	10.20
3	12.50	16.80	14.20	0.60	6.90	0.00	43.90	5.20
4	5.30	24.70	48.40	2.70	0.00	0.00	11.50	7.40
5	0.00	56.20	12.40	0.30	0.00	3.40	24.90	2.90
Average	11.68	20.64	20.86	1.40	8.58	3.52	25.3	8.04

Note: Mud and gravel had been removed from the samples

Moisture contents of the MSW components are summarized in Table 2. As expected, paper, polystyrene, and yard wastes had the highest proportion with 26.7%, 25.0%, and 28.8%, respectively. Rubber had the lowest percentage of moisture with 10.2%. The high moisture content is normally attributed to the climate of the area. Perlis is a tropical climate state with a large amount of rainfall obtained throughout the year. The annual rainfall is approximately 2,700 mm and may increase up to 6,000 mm in the catchment region. On top of this, the average air humidity in Perlis is above 80% [15].

Table 2. Moisture content of MSW components from Padang Siding, Perlis Landfill

Waste Components	Wet weight (kg)	Dry weight (kg)	Moisture Content (%)
Food waste	0.780	0.682	12.6
Paper	0.030	0.022	26.7
Plastic	0.286	0.238	16.8
Polystyrene	0.024	0.018	25.0
Rubber	0.510	0.458	10.2
Yard waste	0.132	0.094	28.8
Wood	0.300	0.268	10.7

Proximate and TGA analysis

Proximate analysis was carried out in order to determine the moisture content (dry basis), volatile matter content, fixed carbon content and ash content in the waste samples. The analysis was conducted for the combination of yard wastes, plastic, paper, polystyrene, wood and rubber. Table 3 tabulates the proximate analysis for 5 samples collected from Padang Siding Landfill. Volatile matter had 68% in average and offered high percentage of condensable gas. The thermochemical conversion method is the best option as condensable gas is promising in amount. The proximate analysis conducted in previous studies set in Penang, Malaysia, shows that the investigated MSW comprised of 44.66% of moisture content, 44.55% of volatile matter, 8.56% of ash content and 2.23% of fixed carbon content [16]. Typically, based on the proximate percentages range for moisture content in residential, MSW is about 15% to 40%, volatile matter comprises approximately 40% to 60% and the range for fixed carbon is from 4% to 15% [17].

Table 3. Proximate analysis of MSW from Padang Siding landfill[a]

Variable (wt. %)	Sample					Average
	1	2	3	4	5	
Moisture content	7	10	5	7	5	7
Volatile matter	53	71	68	71	79	68
Fixed carbon	10	7	10	10	2	8
Ash content	30	12	17	12	14	17

Note: [a] dry basis

The TG analysis is usually used for thermal characterization, calculation of the pyrolysis conversion and evaluation of reactivity and kinetic parameters [7]. In this research, the purpose of TG analysis was to look into the degradation profile for MSW. Figure 1 shows the thermal decomposition of MSW. The initial curve indicates that there was moisture loss of up to 105 °C. The decomposition was gradually increased starting from 200 °C until it had a fast decomposition reaching 350 °C. Up to 400 °C, the remaining residue was 40% and about 60% was decomposed. The decomposition produced approximately 15 to 17% of ash content. This result is in agreement with the waste compositions described earlier. The carbon residues ranged between 10 to 17% pose significant carbon deposition during refinery upgrading (e.g. catalytic cracking), whereas the values above 20% are considered problematic. The decomposition trend was observed to be influenced by the waste compositions, such as paper, wood and plastic. For each TG experiment, the weight loss of MSW as a function of temperature cannot be simulated by considering a single decomposition reaction [18]. The remaining others slowly degraded over 350 °C until 800 °C. They are 4 zones indicated as A, B, C and D in thermogravimetric curve and this is presented clearly in the DTG curve.

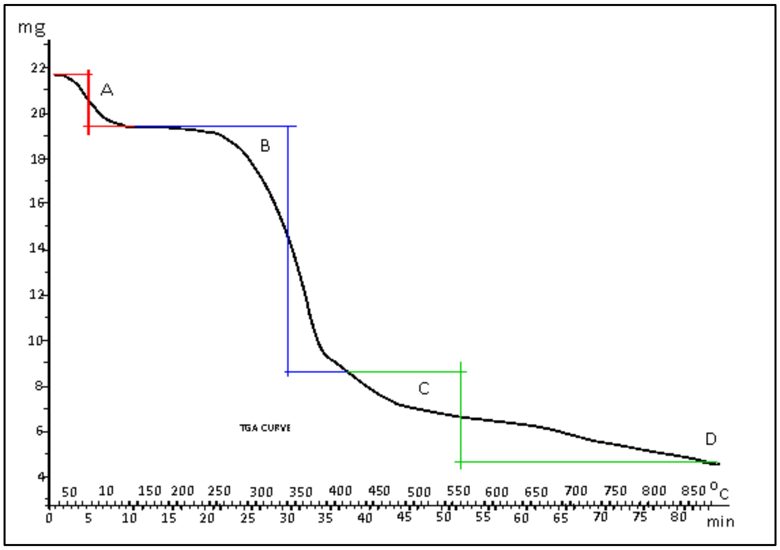

Figure 1. TGA curves for MSW samples

Figure 2 shows the DTG evolution profiles for MSW samples. Zone A indicated moisture loss curve while Zones B, C and D were attributed to the waste compositions. The fast decomposition occurred at three regions which were at 350 °C, 430 °C and 700 °C. The fast decomposition at zone B was caused by the degradation of materials, such as wood and yard wastes. Based on the DTG evolution profiles, material at zone B started to decompose at temperature ranging from 200 °C to about 400 °C, with T_{max} at 350 °C. It was followed by the degradation of plastic and the fast decomposition was shown at C. Zone C recorded the burn-out temperature starting from 400 °C and ended at 480 °C. The more stable materials, such as rubber, gradually degraded and recorded fast decomposition at 700 °C. Small percentage of rubber contributed to the small peak at 700 °C. The burn-out temperature occurred at 600 °C with T_{max} at 750 °C. The differences in the DTG profiles showed the difference in the complexity of MSW components. Shen and Qinlei reveal three steps in their previous study due to the emission and combustion of the volatiles with the char combustion [19].

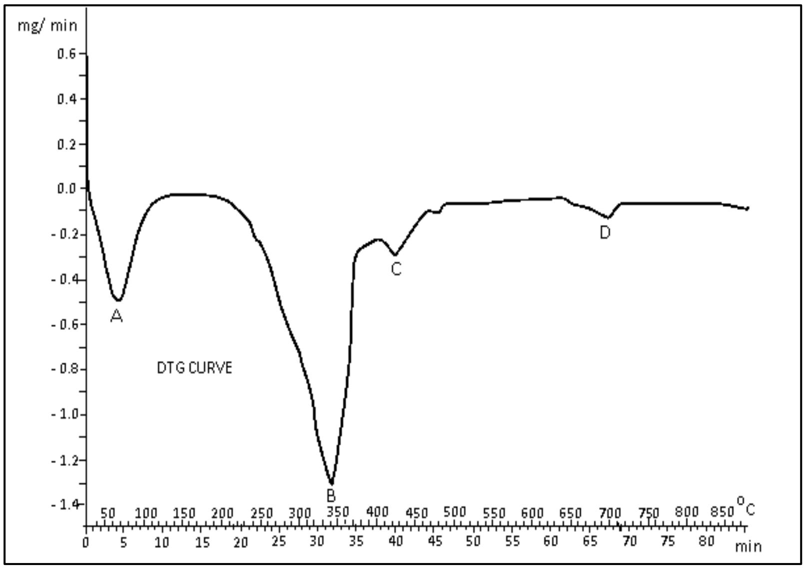

Figure 2. DTG curves for MSW samples

Calorific values (CVs)

Table 4 shows the calorific values of MSW by using bomb calorimeter, calculations employing physical composition and proximate analysis techniques. The calorific values ranging from 2200 to 4500 kcal/kg were obtained for all samples by using bomb calorimeter. Sample No. 4 had the highest value while sample No.1 recorded the lowest. Kathirvale et al. reported that the calorific values of the Malaysian MSW ranged between 1500 and 2600 kcal/kg [20]. The CVs calculated employing physical composition gave values ranging from 1601 to 5550 kcal/kg while proximate analysis technique gave values ranging from 2343 to 4000 kcal/kg. In physical composition, sample No. 4 gave the highest value with 5550 kcal/kg. This was attributed to the composition of paper and plastic and it is in agreement with PL/PA ratio as reported by Abu-Qudais and Abu-Qdais [5]. Traditional method shows a lower error compared to the physical method. The result from traditional method analysis is useful in calculating CVs in MSW study compared to the physical method.

Table 4. Comparison of CVs determined by different methods

Waste Sample	**Calorific Value (kcal/kg)**		
	Bomb Calorimeter	**Physical Composition**	**Traditional Method**
1	2200	1802	2343
2	2500	1601	3135
3	3451	2212	3030
4	4500	5550	4000
5	3480	3717	3525

Thermochemical processes

The pyrolysis experiments were conducted using fixed bed pyrolyser at pyrolysis temperature of 470 ^{o}C and the bio-oil produced was ~ 35%. The hydrous pyrolysis were conducted at 380, 420 and 480 ^{o}C to investigate the effect of temperature on product distribution (Figure 3). The high gas yield obtained at 480 ^{o}C (42%) had resulted in the decrease of liquid products due to the further thermal decomposition [21]. As decomposition to form gas occurred when the temperature had increased to 480 ^{o}C, the oil plus water yield decreased to 42%. Of this, 6% was recovered as heavy oil while approximately 10 – 12% of heavy oil was recovered at 380 and 420 ^{o}C.

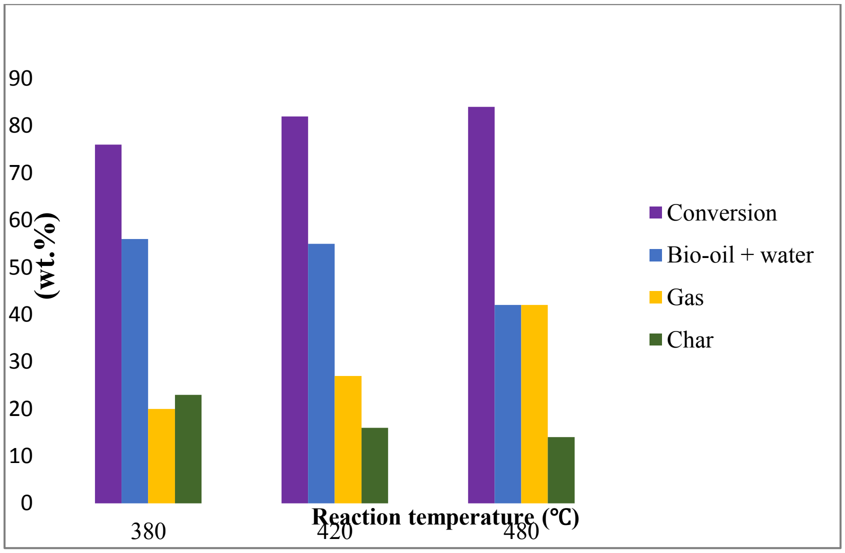

Figure 3. Effect of reaction temperature on product yields using MSW sample to water mass ratio of 1:6 for 1 hour

Table 5 presents the elemental compositions of the bio-oils and heavy oils obtained from pyrolysis and hydrous experiments. An increase in temperature gave low oxygen contents (~11%) for heavy oil from hydrous pyrolysis and bio-oils obtained from direct fast pyrolysis had high oxygen content (~49 – 53%).

Table 5. Elemental analyses of bio-oils and heavy oils from pyrolysis and hydrous pyrolysis experiments

Runs	Samples	C (%)	H (%)	N (%)	*O (%)
Pyrolysis	Bio-oil 1	42.0	7.0	0.3	50.7
	Bio-oil 2	43.0	6.9	0.4	49.7
	Bio-oil 3	40.0	7.0	0.3	52.7
Hydrous pyrolysis	Heavy oil (380 °C)	74.0	6.8	0.6	18.6
	Heavy oil (420 °C)	77.0	7.1	0.6	15.3
	Heavy oil (480 °C)	81.0	7.3	0.5	11.2

*Oxygen by difference

Conclusion

The MSW in Perlis mainly consists of plastic and paper followed by food waste, wood, yard waste and polystyrene. Referring to the wide range of wastes, the thermogravimetric analysis displays a broad range of degradation temperature starting from 200 °C to 800 °C. DTG curves illustrate the fast decompositions that occurred at 350 °C, 430 °C and 700 °C which represent different waste components. For each TG experiment, the weight loss of MSW due to temperature cannot be simulated by considering a single decomposition reaction. CVs calculation employing traditional method gave a lower error compared to the physical method. The physical composition method could be employed for MSW CVs calculation if the MSW compositions are influenced by ratio of plastic to paper. Bio-oil production via fast pyrolysis route gave higher liquid yield of ~35% with high oxygen content of ~49 – 53%. Heavy oil recovered from hydrous pyrolysis experiments gave low yield of ~7 –12%, however with low oxygen content of ~11 – 18%.

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