



Converting Styrofoam Waste into Fuel Using a Sequential Pyrolysis Reactor and Natural Zeolite Catalytic Reformer

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Abstract. Converting 185tyrofoam plastic waste (SPW) into liquid pyrolysis oil (PLO) using a sequential pyrolysis reactor and natural zeolite catalytic vessel was conducted at relatively low temperatures from 160°C to 260°C and complete pyrolysis time, t_p of 60-120 minutes. The presence of natural zeolites on a reformer vessel was intended to undergo the second stage degradation of pyrolysis gas produced without further heating. This work aims to find the yield and quality of PLO that are significantly influenced by the feedstocks of SPW and natural zeolite as a catalyst. The modification of zeolite enhances its fundamental characteristics and its effect on thermochemical behavior and pyrolysis reactions. Utilizing natural zeolite catalysts as TA-ZN (thermal activation) and AA-NZ (acid activation) increases the yield and quality of PLO as fuel. The utilization of the AA-NZ catalyst provided the highest PLO yield, lowest wax, and smallest solid deposit in the shortest at complete pyrolysis time, t_p . PLO produced from SPW pyrolysis with AA-NZ catalyst contains a higher range of hydrocarbon gasoline (C_5 - C_{12}) as styrene, ethylbenzene, and methylbenzene with styrene compound as the highest amount of 74.63% and without a heavy hydrocarbon compound fraction ($>C_{20}$). According to the analysis of all physical properties and GC-MS characterization, this PLO lies within a range comparable to that of conventional commercial kerosene.

Keywords: Catalytic; Fuel; Natural-zeolite; Pyrolysis; Styrofoam

1. Introduction

Recently, plastic waste is one of the largest components of municipal solid waste (MSW) and styrofoam plastic waste (SPW) is the most abundant (Miandad *et al.*, 2017a). In the city of Bandung, West Java, Indonesia, the household and non-household sectors generated 21,769 and 11,951 tons of solid municipal waste per month, respectively (Fitidarini and Damanhuri, 2011). Due to the infeasibility of mechanical (physical) recycling, the majority of SPWs are disposed of in landfills or as litter. SPW can also be recycled through thermal conversion techniques such as incineration, gasification, and pyrolysis. For both environmental and economic reasons, pyrolysis is preferable to incineration and gasification as an option to reduce SPW disposal through landfills (Miandad *et al.*, 2017b). Pyrolysis is a thermal cracking in the absence of oxygen with or without a catalyzer to convert SPW into liquid fuel, char, and gases (Sogancioglu, Yel, and

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Ahmetli, 2017; Suhartono *et al.*, 2018). Pyrolysis is a promising technology for converting SPW to produce liquid oil as an alternative energy source (Rehan *et al.*, 2016).

In the pyrolysis process, catalysts are used to improve the yield and quality of liquid oil products. Various catalytic cracking cracks (FCC), silica-alumina, cellular composition material (CMC), zeolite, etc. are utilized in the waste plastic pyrolysis to increase the yield of oil products (Hossain *et al.*, 2019; Rashid *et al.*, 2019; Suhartono *et al.*, 2018). The significant pyrolysis product yields were obtained from the conversion of various plastic wastes using natural zeolite as a catalyst (Miandad *et al.*, 2017b). The light oil yield increased due to the presence of natural zeolite catalysts in the pyrolysis of plastic waste (Syamsiro *et al.*, 2014). Gasoline and light oil yields significantly increased in the utilization of zeolite socony mobile (ZSM)-5 catalysts in the pyrolysis of plastic waste at 520 and 420°C (Mohanraj, Senthilkumar, and Chandrasekar, 2017). The majority of other researchers hypothesized that natural zeolite could potentially be utilized as a catalyst in the pyrolysis process to convert various plastic wastes into liquid oil. The properties of zeolites such as surface area, pore size, pore volume, and acidity are the main factors that affect their performance as a catalyst. Zeolite crystal size affects the surface area, which has a major impact on porosity, diffusion, adsorption, and chemical reactions. The use of zeolite catalyst increases the rate of the cracking reaction which leads to an increase in the yield of PLO (Nizami *et al.*, 2017; Miandad *et al.*, 2017c).

There are approximately 205.82 million tons of zeolite reserves dispersed across numerous islands in Indonesia. The hypothetical zeolite resource in the Cikembar region of Sukabumi, West Java, is 24,151,000 tons (Sriningsih *et al.*, 2014). This natural zeolite is rarely used as a catalyst for the pyrolysis process. In preliminary experiments, natural zeolite (NZ) and active acid zeolite (AA-NZ) have good adsorption capacities of methylene blue solution of 0.0215 mg/g and 0.061 mg/g, respectively. Therefore, one of the main considerations in this study is to utilize it as an inexpensive catalyst. This research is to study the effect of utilizing natural zeolite as a catalyst to convert SPW into liquid oil, characterizing the composition and properties of liquid oil produced and examining the quality of liquid oil corresponding to a commercial fuel.

2. Methods

In this experiment, two variations of the SPW reduce by 150 g and 300 g were used as feedstock, which was fed to the pyrolizer reactor for degradation and then continued with the second stage degradation on the catalytic reformer vessel. Zeolite as a catalyst is made from zeolite rocks with an average size of 6 cm obtained from the Cikembar area, Sukabumi, West Java province of Indonesia. To increase its effectiveness, this zeolite must first be prepared to increase its surface area (Sudibandriyo and Putri, 2020). Due to the difficulty and high cost of recovering (regenerating) the catalyst, both unmodified and modified natural zeolites were applied once as a catalyst for SPW pyrolysis. For each type of solid natural zeolite catalyst, as much as 600 g was applied indirectly with SPW to the pyrolysis reactor by inserting it into the reformer reactor using a perforated bucket.

Gas chromatography-mass spectrometry (GC-MS) was utilized to analyze the chemical composition (fraction) of liquid oil products. Some significant physical and combustion properties of liquid oil produced were evaluated to meet the hydrocarbon fuel range, according to the procedure established by American Standard Testing and Material (ASTM).

2.1. Feedstock and catalyst preparation

The Styrofoam Plastic Waste (SPW) was collected from trash cans around the campus. For every 2 kg/day of plastic waste, 50% of which is a type of styrofoam plastic. As depicted in Figure 1, this SPW was cleaned and dried conventionally, then cut into small pieces measuring approximately 2 cm².

In a previous study, the researchers discovered that using a natural zeolite catalyst with a particle size of less than 10 mm induced blockages in the catalytic reformer reactor and cooling piping due to condensable gas produced during pyrolysis (Suhartono, *et al.*, 2018). Therefore, a solid natural zeolite of about 10 mm in size was utilized for the second stage degradation of SPW. Three types of natural zeolite catalysts were prepared, namely natural zeolite (NZ), temperature-modified natural zeolite catalyst (TA-NZ), and acid activation-modified natural catalyst (AA-NZ). Natural zeolite (NZ) was physically prepared by crushing to the size of a granule of less than 10 mm.



Figure 1 Styrofoam plastic waste as pyrolysis feedstock

The washed NZ was dried and calcined at 120°C and 500°C, respectively, and then so call TA-NZ. To improve its catalytic characteristics, TA-NZ was used for the preparation of chemically modified catalysts through acid activation. TA-NZ was immersed in a 1% (v/v) HF solution and then submerged in 1 M HCl for 24 hours. After being filtered and washed, TA-NZ was dried in an oven at 130°C for 3 hours. The grain catalyst was then soaked with NaOH solution for 24 hours and followed by drying in an oven at 120 °C to get a modification of catalytic acid activation, namely AA-NZ (acid activation-NZ).

2.2. Experimental setup and scheme

A small pilot-scale pyrolysis reactor was constructed and utilized to convert SPW into liquid fuel as the main product. As demonstrated in Figure 2, the equipment arrangement consists of the following main parts: I pyrolysis reactor, (ii) glass wool insulation room, (iii) catalytic removal, (iv) LPG heating system, and (v) condenser cooling system.

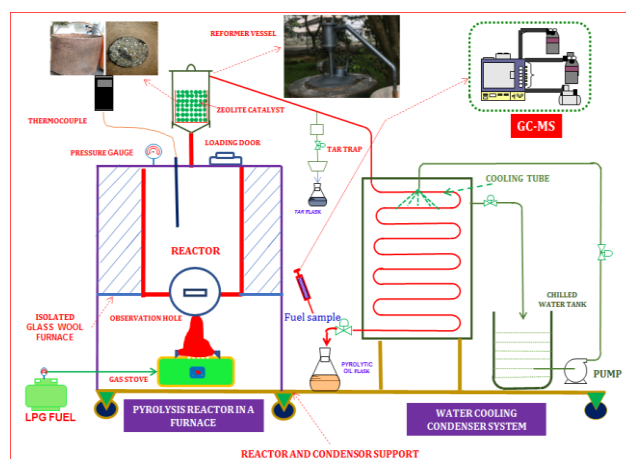


Figure 2 A sequential pyrolysis reactor and reformer

The Pyrolysis reactor with an inner diameter and height of 255 mm and 328 mm respectively was made of stainless steel and isolated using glass wool in a chamber. The pyrolysis reactor was heated up to a pyrolysis temperature of 260°C. K-type (chromel alumel) thermocouple was inserted exactly into the pyrolysis zone inside the reactor. This reactor was equipped with a perforated bucket and connected to a 1.5 L catalytic reformer. The second stage degradation of these volatile vapors occurred in this catalytic reformer without any further heating. The cooling tube length of 2500 mm was immersed in a circulated water chiller tank with a height and diameter of 700 mm and 500 mm, respectively. The volatile vapors were cooled in the condenser system to obtain the liquid product as fuel. The solid product as remaining carbon and residue were also observed.

2.3. Pyrolysis liquid oil characterization

The chemical composition (fraction) of liquid oil products was analyzed by using gas chromatography-mass spectrometry (GC-MS, HP 7890 with a 5975-quadrupole detector). The mass spectra on the retention time of the unknown chemical compounds. By using the National Institute Standard and Technology (NIST) data repository, researchers were able to infer the identities of the unknown chemical compounds from their chromatographic peaks (Stein and Wallace, 2017). The mass percentage of different chemical compounds in liquid oil produced was appraised as the percentages of the peaks to the total ion chromatogram (TIC) peak area.

Some significant physical and combustion properties of liquid oil produced were evaluated to meet the hydrocarbon fuel range. Following the American Standard Testing and Materials (ASTM) D1234 methods, the volumetric glassware was used to measure the density of liquid oil at 30°C in three times repeatable to ensure accuracy. A Cannon-Fenske capillary viscometer was used to determine the viscosity of liquid oil using the ASTM D445 method. In the meantime, the heating value of liquid oil was determined using equations that are commonly used because of their accuracy and affordability. These equations were based on measurements of the oil's density and viscosity (Suhartono *et al.*, 2018; Demirbas, 2000). The standard test method of the Cleveland open-cup tester (ASTM D92-05) and tag open-cup apparatus (ASTM D 1310) was used to determine the flash point and fire (ignition) point of the PLO (ASTM, 2005; Suhartono, 2019). The water boiling test method (WTB) was adopted to evaluate the pyrolysis liquid oil as an alternative energy source using a conventional wick stove to replace kerosene fuel by calculating the fuel consumption (Suhartono, Putri, and Fauziah, 2017; Visser, 2015). The flame temperature was observed using probe thermometry by placing it into the surface flame.

3. Results and Discussion

3.1. Pyrolysis product yields

SPW pyrolysis can be carried out at relatively low temperatures ranging from 160°C to 260°C with a gradual increase in temperature of 0.8°C/min. at a pressure slightly higher than atmospheric pressure (± 1.2 atm). As shown in Table 1 and Figure 3, the total amount of PLO was obtained at temperatures of 215°C, 244°C, 230°C, 260°C, 225°C, and 235°C for 60 min., 80 min., 120 min., 70 min., and min., respectively. This is in line with previous research reports that SPW can be converted into liquid fuel at a low temperature of 150°C-300°C and a processing time of 20-60 minutes in the fixed bed type reactor (Kholidah, Faizal, and Said, 2019).

According to Table 1, the collected pyrolysis liquid oil was divided into two phases: the liquid fuel oil phase and the wax phase (paraffin). The minuscule solid deposit (coke

and residue) was collected, while non-condensable gas products were released into the atmosphere. The yield of liquid fuel oil products is measured by comparing the volume of oil to the weight of the SPW feed, as illustrated in Figure 3.

Table 1 The number of products from pyrolysis SPW

Catalyst	SPW (g)	Pyrolysis liquid oil (mL)		Solid deposit (g)	t_p (min.)
		Fuel	Wax		
Without	150	58	33	0	60
	300	158	29	3.13	80
TA-NZ	150	72	35	0	80
	300	198	26	0.97	120
AA-NZ	150	95	20	0	70
	300	220	18	0.88	90

The presence of natural zeolite as a catalyst has a substantial effect on the number of products produced during the conversion of SPW to PLO, as demonstrated in the table above (Yuliansyah *et al.*, 2015). The complete pyrolysis time, t_p affects the amount of liquid pyrolysis oil produced, the longer the pyrolysis process lasts, the more PLO is produced. The lower solid deposits are due to the use of catalysts in SPW pyrolysis (Kholidah, Faizal, and Said, 2019). A slightly higher difference in the pyrolytic oil was produced due to the utilization of the AA-NZ compared to the TA-NZ. The previous studies also revealed that the use of natural zeolites in the pyrolysis of plastic wastes increased the higher PLO (Suhartono *et al.*, 2018). This result was consistent with the results of a previous study which concluded that the utilization of natural zeolite in the pyrolysis of styrofoam produced higher PLO compared to the use of synthetic zeolites of HZSM-5 (Miandad *et al.*, 2017b).

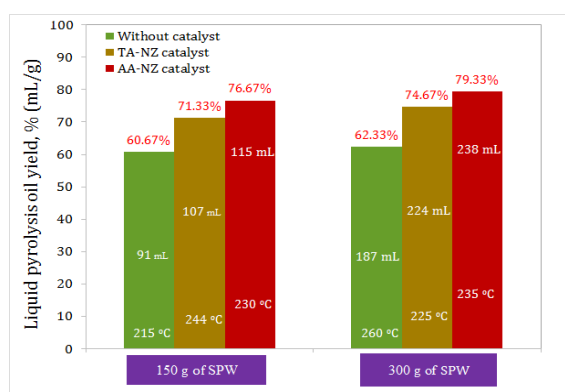


Figure 3 Percentage of pyrolysis liquid oil

Figure 3 demonstrates that the usage of AA-NZ catalyst at 300g of SPW pyrolysis provided the highest fuel oil of 219 mL, the lowest wax of 18 mL, and the smallest solid deposit of 0.88 g in the shortest at complete pyrolysis time, t_p of 90 minutes. In terms of comparisons, SPW with zeolite catalyst at 275°C and SPW without zeolite catalyst at 250°C resulted in average pyrolysis oil outputs of 48.6% and 47.9%, respectively (Shah *et al.*, 2010). In the pyrolysis process, the presence of a catalyst will reduce the activation energy and increase the reaction rate (Jamilatun *et al.*, 2020). The TA-NZ catalyst produced slightly less pyrolysis oil than the AA-NZ catalyst due to acid activation, which increases surface contact with pyrolysis vapors (Syamsiro *et al.*, 2014; Miandad, *et al.*, 2016). The presence of natural zeolite catalysts enhances the degradation of long-chain (heavy) hydrocarbons from pyrolysis vapors into lighter hydrocarbons in a catalytic reformer. The presence of natural zeolite catalysts changes the composition of PLO into

the gasoline hydrocarbon range (C_5 - C_{12}) (Mohanraj, Senthilkumar, and Chandrasekar, 2017). In this work, the use of zeolite catalysts decreased the complete pyrolysis time, t_p , resulting in a decrease in energy consumption.

Figure 3 above depicts a higher percentage of liquid oil yield of 80% which was also obtained when using TA-NZ (600 g) catalyst at a very close pyrolysis temperature but a longer degradation time of 120 minutes. The slightly higher percentage of liquid pyrolysis oil yield of 79.33% was obtained from 300 g degradation of SPW at a pyrolysis temperature of 250°C and a complete pyrolysis time of 90 minutes using AA-NZ (600 g) catalyst. A similar phenomenon was also found in the pyrolysis of a different amount of SPW 150 g. The previous result reported that liquid yields of 85-95% were found from degraded 50 g SPW at 500°C in 2 hours using the catalytic reformer, in the presence and absence of the natural zeolite (Syamsiro *et al.*, 2014). Pyrolysis of SPW in sequence with the second-stage degradation using the AA-ZN catalyst produced the highest yield of pyrolysis liquid oil compared to TA-NZ and without the catalyst. This was due to the use of AA-NZ which facilitates the degradation of pyrolysis organic vapors into lighter hydrocarbon compounds, followed by effective condensation in large condenser systems. Compared to the TA-NZ and NZ catalysts, the modified AA-NZ catalyst induced a greater catalytic degradation of gaseous products due to its greater surface area, size, pore volume, and acidity (Miandad *et al.*, 2017b). In this case, it seems that a higher pyrolysis temperature was unnecessary, although it could increase gas products, some of which were incondensable hydrocarbon gas chains (Kholidah, Faizal, and Said, 2019). The shortest complete pyrolysis time, t_p was observed when AA-NZ was used to degrade the organic vapor in the pyrolysis of SPW, as shown in Table 1. The shorter the time pyrolysis is required the less fuel consumption is used for the heating process.

3.2. GC-MS analysis of pyrolysis liquid oil

The results of GC-MS of PLO using AA-NZ catalyst are summarized in Table 2. Styrene as a mono-aromatic compound represented the largest percentage of 86.47% compared to others. The single aromatic ring compound was also present in significant amounts of liquid pyrolysis oil. Polyaromatic ring compounds such as naphthalene (1.688%), dibenzyl ethanol (0.888%), and 1,3 propanediol benzene (0.364%) were found as the smallest percentage.

The GC-MS results indicate that the liquid pyrolysis oil produced by SPW pyrolysis with the AA-NZ catalyst produced contains a higher hydrocarbon gasoline range (C_5 - C_{12}) compared to other hydrocarbon fractions. The composition of the liquid oil consists mainly of styrene, ethylbenzene, and methylbenzene in the gasoline hydrocarbon fraction (C_5 - C_{12}) also stated by peer researchers (Syamsiro *et al.*, 2014). These findings suggest that liquid pyrolysis oil products still contain styrene in the majority of cases.

Table 2 GC-MS results of pyrolysis liquid oil from pyrolysis of SPW with AA-ZN catalyst

R. time	Peak	Compound name	Fraction (%)	Molecular formula
3.629	527564	Methyl benzene	5.529	C_7H_8
5.022	216458	Ethyl benzene	1.942	C_8H_{10}
5.495	8960634	Styrene	86.478	C_8H_8
6.981	211339	1-methylethenyl benzene	3.111	C_9H_{10}
16.820	185241	1,3 propanediyl benzene	0.364	$C_{15}H_{15}$
17.402	898334	1,2,3,4 tetrahydro-2-phenyl naphthalene	1.688	$C_{16}H_{16}$
22.374	555530	Dibenzyl ethanol	0.888	$C_{16}H_{18}O$

TA-NZ catalysts result in a slightly different liquid oil pyrolysis product composition than AA-ZN. Both catalysts produce PLO, which is mostly comprised of aromatic chemicals

with styrene as the primary component. As can be seen in Table 3, the percentage of styrene compounds is highest with the TA-NZ catalyst (74.625%), followed by the AA-NZ catalyst (86.478%).

Table 3 GC-MS results of pyrolysis liquid oil from pyrolysis of SPW with TA-NZ catalyst

R. time	Peak	Compound name	Fraction (%)	Molecular formula
3,635	527564	Toluene	4.245	C ₇ H ₈
5,022	216458	Ethyl benzene	2.019	C ₈ H ₁₀
5,489	8960634	Styrene	75.624	C ₈ H ₈
6,987	211339	Methyl ethyl benzene	3.140	C ₉ H ₁₀
16,814	185241	1,3 propanediol	1.974	C ₁₅ H ₁₆
17,397	898334	1,2,3,4 tetrahydro-2-phenyl naphthalene	8.069	C ₁₆ H ₁₆
22,374	555530	2,3 diphenyl cyclopropyl	4.928	C ₂₂ H ₂₀ OS

The use of TA-NZ produces a slightly lower gasoline hydrocarbon fraction (C₅-C₁₂) than styrene, ethylbenzene, and methylbenzene. Using the TA-NZ catalyst instead of the AA-NZ catalyst resulted in slightly higher kerosene (C₁₁-C₁₄) and diesel hydrocarbon fraction (C₁₁-C₁₈) (Prihadiyono *et al.*, 2022). The heavy oil fraction (>C₂₀) also appeared with the use of the TA-NZ catalyst, while the use of the AA-ZN catalyst did not produce a heavy hydrocarbon compound fraction (> C₂₀). The total fraction of light hydrocarbons of 97.06% and 85.02% were produced from the use of TA-NZ and AA-NZ, respectively. Thus, it can be stated that the pyrolysis oil produced using a catalyst is in the range of the hydrocarbon gasoline fraction (C₅-C₁₂).

3.3. Pyrolysis liquid oil (PLO) properties as a fuel

As summarized in Table 4, the purpose of the SPW PLO product characterization was to demonstrate that the product can be used as a fuel comparable to conventional. The PLO density was found to be exactly within the range of kerosene density. The density is closely related to molecular weight; the higher the hydrocarbon molecular weight means the higher the density. Additionally, the specific gravity indicates energy density too. Therefore, in this case, the same density must indicate that the molecule has identical properties. Density is one of the PLO's physical properties as a fuel that is more responsible for combustion performance. Higher fuel density results in lower volatility and poor atomization of PLO during fuel injection in the combustion chamber which causes incomplete combustion and carbon deposits. The average value of PLO density (0.768-0.890 g/cm³) produced from the pyrolysis of SPW with and without zeolite catalysts is in the range of the value of kerosene density as listed in Table 4. The PLO density of the AA-NZ and TA-NZ pyrolysis of 0.768 g/cm³ and 0.812 g/cm³, respectively was found to be exactly within the range of kerosene density. This may be due to the high aromatic hydrocarbon content as a light oil in the gasoline fraction (C₅-C₁₂) compared to non-catalyst PLOs. In its application, the high density of fuel results in decreased volatility and poor atomization, whereas the lower density of fuel oil will have a greater impact on fuel consumption and vice versa (Suhartono *et al.*, 2018).

The slightly lower viscosity of PLO than kerosene is related to the light oil in the gasoline fraction containing aromatic hydrocarbons (C₅-C₁₂). Conversely, the higher viscosity of PLO than kerosene is due to a slightly higher content of kerosene hydrocarbon fraction (C₁₁-C₁₄) and diesel hydrocarbon fraction (C₁₁-C₁₈). Viscosity is influenced by molecular weight; the greater the molecular weight of a hydrocarbon, the greater its effect on the viscosity of the fuel (Mohammed, Attiya, and Khudai, 2008). The double bonds stretching of aromatic hydrocarbon compounds in the PLO induce to decrease in the oil

viscosity (Suhartono, Putri, and Fauziah, 2017). The viscosity of fuels plays an important role in the combustion process. The higher viscosity of PLO leads to poor atomization that causes incomplete combustion. The kinematic viscosity of the PLO is slightly lower than the range of commercial kerosene fuels, so it does not require further reduction to be used as a fuel on par with kerosene for domestic purposes.

Table 4 Pyrolysis liquid oil (PLO) and conventional fuel properties

Parameter	Pyrolysis liquid oil			Conventional fuel		
	Noncatalytic	TA-NZ	AA-NZ	Gasoline	Kerosene	Diesel
Density, g/cm ³	0.890	0.812	0.768	0.715-0.780	0.775-0.840	0.875-0.959
Viscosity, cSt _(30°C)	2.221	1.944	1.663	0.40-0.88	2.4-2.71	2.00-3.27
Flashpoint, °C	33	35	37	-(23-43)	38-41	52-82
Autoignition point, °C	248	225	218	247-280	220-295	254-285
HHV, MJ/kg	41.73	41.15	41.03	43.1-46.2	44-46	42-46

The PLO flashpoint was observed at 33-37°C, which is lower than the flashpoint range of kerosene at 38-41°C. As a result of its lower flashpoint, PLO is a more flammable fuel than kerosene. The PLO autoignition range of 218-248°C is within the kerosene automatic ignition range of 220-295°C. This implied that the PLO will be as flammable as kerosene fuel. The HHV value of PLO of about 41 MJ/kg was produced from PSW pyrolysis both without catalyst, with TA-NZ and AA-NZ catalysts. The PLO HHV value is slightly lower than other fuels due to the predominance of light hydrocarbon compounds by the decrease in hydrogen content. However, this HHV value corresponds to the HHV of 41.6 MJ/kg obtained from the pyrolysis of PSW using a small-scale pyrolysis reactor at 400°C and 75 minutes was also revealed (Miandad *et al.*, 2016).

3.4. Potential application of PLO as a fuel

PLO from pyrolysis of SPW containing large amounts of mono and poly-aromatic ring compounds with different fractions has the potential to be utilized as a source of energy. PLO produced with the largest content of a single aromatic ring of styrene compound can be further treated to upgrade its quality to the gasoline hydrocarbon range. The combustion of PLO as fuel was tested using a conventional wick stove. Thermal efficiency is used to evaluate the performance of a PLO, stove, and pot combination by adopting the Water Boiling Test (WBT) method within 10 minutes of operation. Average thermal efficiency of around 28.30 -58.10% by the fuel consumption rate (FCR) of 0.10-0.18 L/h. In comparison, the thermal efficiency of the two types of kerosene-fueled wick stoves is 55% and 46%, respectively (Makonese *et al.*, 2012). The heating value of PLO of 43.83 MJ/kg is produced from polystyrene in a fixed-bed reactor provided by an electric heater which can be used as a fuel for other comparisons (Abdullah *et al.*, 2018).

4. Conclusions

Based on the analysis of all characteristics and physical properties, the PLO produced is comparable to conventional commercial kerosene. Domestically, the PLO can be utilized as a direct substitute for kerosene fuel. Utilization of this PLO as fuel using conventional wick stoves provides a thermal efficiency of around 28.30-58.10% with a fuel consumption rate (FCR) of 0.10-0.18 L/hr.

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