Microwave-assisted pyrolysis of plastic waste for liquid oil production

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ABSTRACT

In this work, the article presents the results of the production of fuel oil from plastic waste using microwave-assisted pyrolysis. Under the influence of microwave power, microwave absorption material, time, catalyst, polyethylene (PE), polypropylene (PP), polystyrene (PS) and mixture (34% PE + 33% PP + 33 wt% PS) were pyrolyzed to liquid oil. The results showed that with 500 g of raw plastic, 500 W microwave power, and 120 minutes, the carbon absorber accounted for 10 wt% (compared to plastic), catalysts accounted for 20 wt%, fuel efficiency reached 84% with PS and PP, 83% with PE, and 83.5% with mixed plastics. The oil obtained qualified the standards of FO No. 1 TCVN 6239:2019. The gas chromatography/mass spectroscopy GC/MS analysis (GC-MS) showed that the composition of the pyrolysis oil is mainly hydrocarbon from C7-C12.

Keywords: Plastic waste; Pyrolysis; Microwave; Oil.

1. INTRODUCTION

Currently, the issue of plastic trash pollution is of the highest priority importance due to its significant impact on both the ecological environment and human well-being [1]. Also, the technology for recycling plastic waste is not only feasible but also cost-effective. So converting plastic waste into a viable supply of liquid fuel is both scientifically and practically beneficial for safeguarding the environment against pollution. Expanding industries are result in limited fossil fuel supplies, such as oil and coal. A number of scientists with an intense interest have conducted research on the utilization of plastic waste for the production of liquid fuels [2].

Recent studies have reported on the pyrolysis techniques of polymers, which include factors such as pyrolysis temperature, residence time, and heating rate [3]. In the pyrolysis temperature range of 250–500 ^{o}C, the oil efficiency can reach 57–70 wt%. As the pyrolysis temperature rises, the oil yield rises because the polymers break down more quickly, and many small-molecule compounds are made [4].

With the further development of pyrolysis technology, microwave-assisted pyrolysis (MAP) has attracted a substantial amount of attention. Researchers have found that this method overcomes some of the limitations of traditional pyrolysis, including slow and uneven heating. The effects of pyrolysis temperature, microwave power, microwave adsorbent, and pyrolysis catalysts on the MAP oil yields and components are investigated [5].

This work presents the results of fuel oil obtained from MAP experiments and the components of PP, PE, PS, and mix oil were tested by the GC-MS method. The effects of microwave power, time, and catalyst loading on the yields of liquid oil were analyze and MAP conditions were discussed for optimal oil yields.

2. EXPERIMENT

2.1. Materials

- Plastic waste collected from Nam Son landfill (Soc Son, Hanoi) and Nghi Yen (Nghi Loc, Nghe An) and then classified, cleaned, cut into 1-3 mm pieces. The obtained plastic waste are the

raw materials chosen to make liquid fuel.

- Carbon black (Macklin) is used as a microwave absorber.
- NiO/ZSM-5 is used as a catalyst.
- N_2 gas (99,99%).

2.2. Experimental system

Figure 1 shows the schematic diagram of a sustainable oil production experimental set-up. The microwave equipment can be set within 0÷1000 W; the frequency was 2450 MHz. The reactor was made of steel with a volume of 1000 mL.

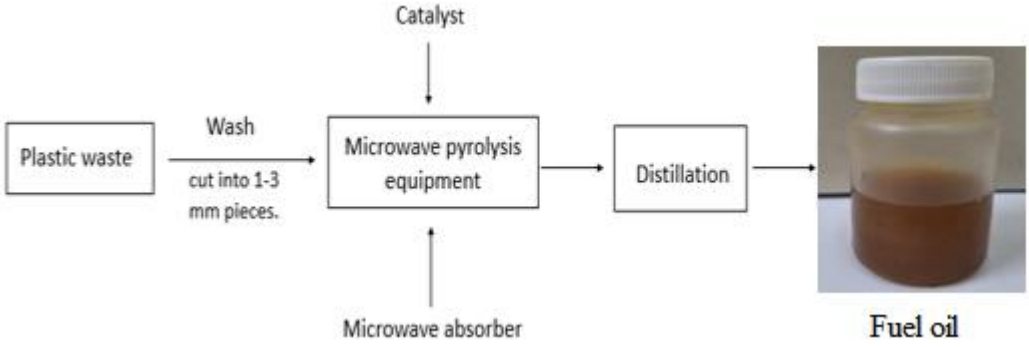

***Figure 1**. Schematic diagram of oil production experimental set-up.*

2.3. Experimental procedures

This study aims to investigate the impact of microwave power, pyrolysis time, carbon absorber content, and catalyst content on oil production efficiency. In the reaction chamber, add 500 g of cut-up plastic waste (PE, PP, PS, or a mixture) and 100 g of carbon-microwave absorber. Make sure to thoroughly combine the mixture. In the catalyst container, add 50 g of ZSM-5 pyrolysis catalyst. To discharge the oxygen in the system, N_2 gas was blown into the reaction chamber at a flow rate of 0.5 L/min. Turn on the microwave to react for 90 minutes; the power consumption is 500 W. Under the influence of temperature, plastics pyrolyzed into lighter products. The liquid-product mixture undergoes distillation to extract oil. The device undergoes cooling and cleaning after the reactions conclude. The formula calculates the efficiency of converting waste plastic into fuel oil.

$$Efficiency = \frac{oil\ weight}{plasitc\ weight} \times 100\% \quad (2.1)$$

Microwave power was set between 300 and 900 W, the microwave absorber and catalyst NiO/ZSM-5 pyrolysis loading were set within 0–20 wt% of the plastic, and the timer was set from 30 to 150 minutes.

We analyze pyrolysis oil products with optimal synthesis conditions to evaluate their composition and properties. The TCVN 6239:2019 establishes the technical specifications and testing techniques for fuel oil furnaces. We used GC-MS analysis (QP 2020 Shimadzu) to determine the composition of the microwave pyrolysis oil. The results provide information on the distribution of carbon numbers in the pyrolysis oil, as well as the distribution of alkanes, alkenes, cycloalkanes and aromatic hydrocarbons.

3. RESULTS AND DISCUSSION

3.1. Effects of microwave power

Temperature plays a significant role in the conversion of plastic into liquid fuel. Liquid oil

conversion efficiency is calculated by the amount of oil obtained compared to the original plastic mass. The temperature of the reactor closely relates to the microwave power. In this study, the microwave power ranges from 300 to 900 W, and corresponding to each power value, the surface temperature of the material is assessed through a thermal sensor. The results are shown in table 1.

Table 1. *The surface temperature of the material at variable power levels.*

Power (W)	Temperature (°C)			
	PE	PP	PS	Mixture
300	343	347	331	340
350	410	405	398	402
400	465	460	450	462
450	480	485	478	482
500	489	492	483	487
550	499	495	492	494
600	502	509	501	505
650	510	521	505	515
700	528	528	516	522
750	535	537	523	533
800	546	549	530	542
850	560	555	540	550
900	572	560	546	565

The results in table 1 show that the microwave power increases from 300 to 600 W, and the surface temperatures of the PE, PP, PS and mixed plastics increase from 330 to 570 °C, respectively. Fuel oil yield with different microwave power values is shown in figure 2.

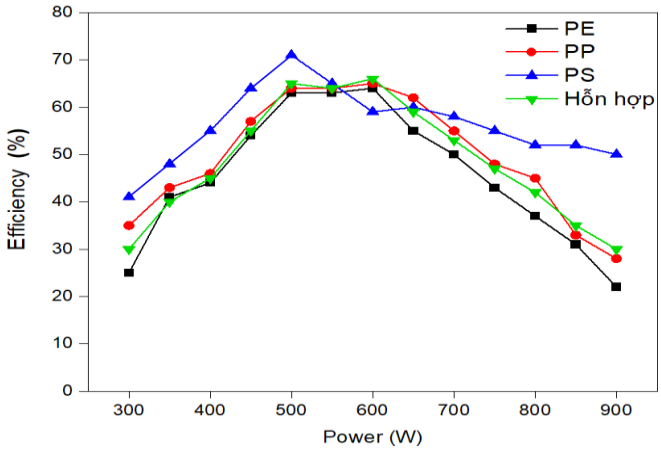

Figure 2. *Fuel oil generating performance at variable power levels.*

As shown in figure 2, the effect of microwave power on the fuel oil yield of PE, PP, PS, and mixed plastics tends to be similar. At 300 to 450 W, the temperature is lower 480 °C, the yield of fuel oil reaches about 50%, and the product contains an amount of wax. Low temperatures prevented the breaking of plastic due to insufficient energy supply, leading to a low oil yield [6]. The maximum fuel oil yield of PS plastics reaches 70% in the 500 to 600 W power range, corresponding to a material surface temperature of 480 to 580 °C, while PE and PS reach 65 and 64%, respectively. As the microwave power increases to 900 W, the liquid-fuel yield decreases significantly. The cracking reaction occurs intensely at high temperatures, which usually generates more gases. This suggests that when the pyrolysis temperature exceeded 550 °C, microwave-

assisted pyrolysis of plastic waste products consisted only of liquid oil and gas. PS is an exception because at high temperatures the main product obtained is still liquid styrene. There was no wax substance in the products, indicating that high pyrolysis power caused all of the plastic macromolecules to crack into small molecules.

3.2. Effects of the amount of microwave absorbent

The amount of microwave absorbent has an influence on the temperature of the surface plastic material and the performance of liquid fuel during pyrolysis. In this experiment, the amount of carbon microwave absorbent was 5%, 10%, 15%, 20%, 25%, and 30 wt% compared to plastic waste. The microwave power was 500 W. Table 2 shows the surface temperature of the material as the microwave absorbent content changes.

Table 2. *The effect of the microwave absorbent content on the surface temperature*.

Cacbon loading (wt%)	Temperature (°C)			
	PE	PP	PS c	Mixture
5	415	410	407	411
10	435	430	436	432
15	467	463	450	460
20	489	492	483	490
25	497	498	485	493
30	502	501	488	500

According to table 2, the microwave absorbent has increased from 5 to 30 wt%, and the surface temperature of plastics is also rising. The microwave absorbent content is 5 wt%, and the surface temperature of the sample reached 405–415 °C. The amount of microwave absorbent is 20%, and the surface temperature of the sample reached about 490 °C. When the weight of microwave absorbent increased, there was a negligible increase in the surface temperature of the sample. Fuel oil yield with different microwave absorbent content values is shown in figure 3.

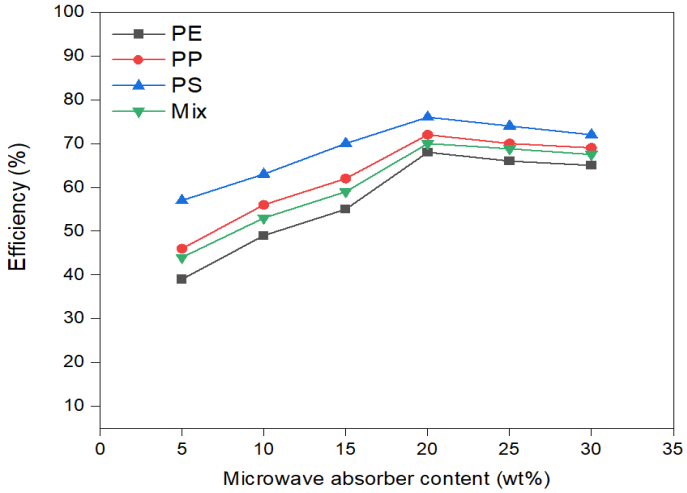

Figure 3. *Fuel oil generating performance at different microwave absorbent content levels*.

Figure 3 shows that fuel oil efficiency tends to increase as the microwave absorbent level increases from 5 to 20 wt%. The microwave absorbent content is 5%, the surface temperature of the plastic material is low, and the plastic pyrolysis reaction occurs slowly. As a result, the liquid yield of PE, PP, and mixed plastics is only about 50%, while PS achieve nearly 60%. The microwave absorbent content at 20 wt%, the liquid yield reaches 76% with PS and approximately

70% with PE, PP, and plastic mixtures. With a higher carbon content, the fuel oil yield tends to decrease slightly. Therefore, we choose the subject containing 20 wt% of the microwave absorber.

3.3. Effects of time

In this study, the duration of thermolysis varies from 15 to 150 minutes. The result present in figure 4.

Figure 4 shows that among plastics, PE exhibits the slowest rate of thermolysis, whereas PS demonstrates the highest rate of pyrolysis under the same conditions. In the first 30 minutes, the presence of thermoplastic is minimal, resulting in a liquid fuel efficiency of less than 10 percent. After 60 minutes, liquid fuel generation has increased more than three times. Between 60 and 90 minutes, the thermolysis reaction intensifies, leading to a rapid increase in performance. After a duration of 120 minutes, the fuel oil efficiency of PS reached 75%. However, with an extended pyrolysis period, the efficiency showed little growth. PP, PS, and mixed plastics exhibit a liquid fuel metabolic efficiency of approximately 70%. Due to the increased reaction time, the plastic undergoes thermolysis, which no longer increases liquid fuel metabolism. Therefore, we have chosen thermolysis reaction times of 120 minutes.

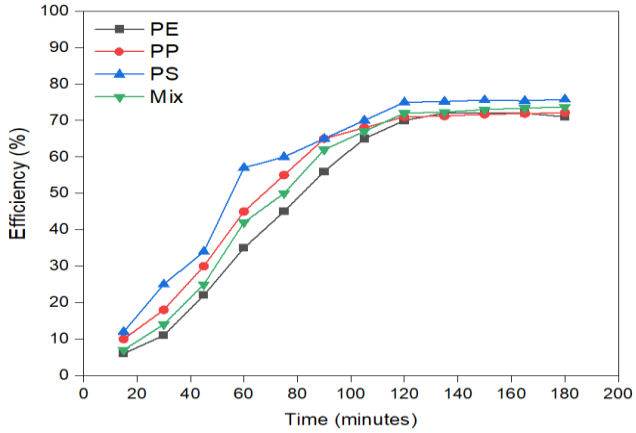

Figure 4. *Fuel oil generating performance at different time.*

3.4. Effects of catalyst

Effects of catalysts were investigated in the range of 0-20 wt%, the result is shown in figure 5.

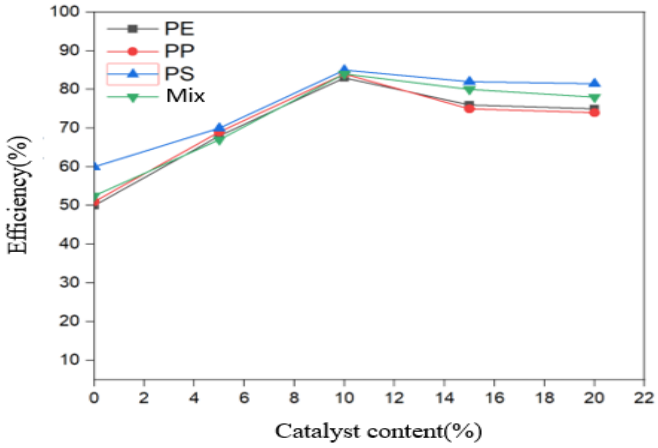

Figure 5. *Fuel oil generating performance at at different catalyst content levels.*

The results in figure 5 show that the catalyst content has a role to play in improving the efficiency of the conversion of plastic waste into fuel oil. In the absence of a catalyst, the efficiency

reaches 60% with PS and approximately 50% with PP, PE, and mixture, resulting primarily in liquid form, although some remains in waxes and gases. When catalyst content increases from 0÷10 wt%, fuel oil yield increases. Using a 5 wt% catalyst significantly increased the efficiency of fuel oil. At a catalyst concentration of 10 wt%, the yield of PS, PP reached 84%, that of PE reached 83%, and the yield of mixture plastics reached 83.5%. By continuously increasing the catalyst content, the fluid oil efficiency decreases. NiO/ZSM-5 catalyst promoted the cracking process, macro-molecules cutting to small molecule. The catalyst content is high, and the cracking reaction happens quickly, creating C1–C4 gas products [7]. From the results we obtained, we selected the catalyst content using plastic waste heat separation to be 10 wt%.

3.5. Fuel oil characterisics

The obtained fuel oil was tested based on standard specification TCVN 6239:2019, including: kinematic viscosity, content of sulfur, ash, residue, water and impurities, heat of combustion, pour point, flash point and specific weight. Results showed that all properties are conformed to the limiting requirements (table 3).

Table 3. *Properties of fuel oil.*

No	Properties	FO oil	Fuel oil			
			PE	PS	PP	Mixture
1	Kinematic viscosity, 50^{o}C, cSt	≤ 87	1,917	1,497	0,994	1,382
2	Sulphur content, wt%	≤ 0,5	0,041	0,036	0,038	0,0061
3	Ash content, wt%	≤ 0,15	0,013	< 0,01	< 0,01	0,030
4	Residue content Conradson, wt%	≤ 6	0,01	0,03	0,02	0,03
5	Water content, % volume	≤ 1	0,0063	0,008	0,021	0,0437
6	Mechanical impurities, wt%	≤ 0,15	< 0,01	< 0,01	0,01	< 0,01
7	Heat of combustion, cal/g	≥ 9800	10993	11074	10261	10671
8	Pour point, ^{o}C	≤ 42	9	-27	-48	9
9	Flash point, closed cup, ^{o}C	≥ 66	68	70	68	67
10	Specific weight 15 ^{o}C, kg/L	≤ 0,991	0,7937	0,7787	0,8839	0,8279

The chemical composition of fuel oil from PE, PS, PP and mixture plastic was determined by the GC-MS method, resulting in the following:

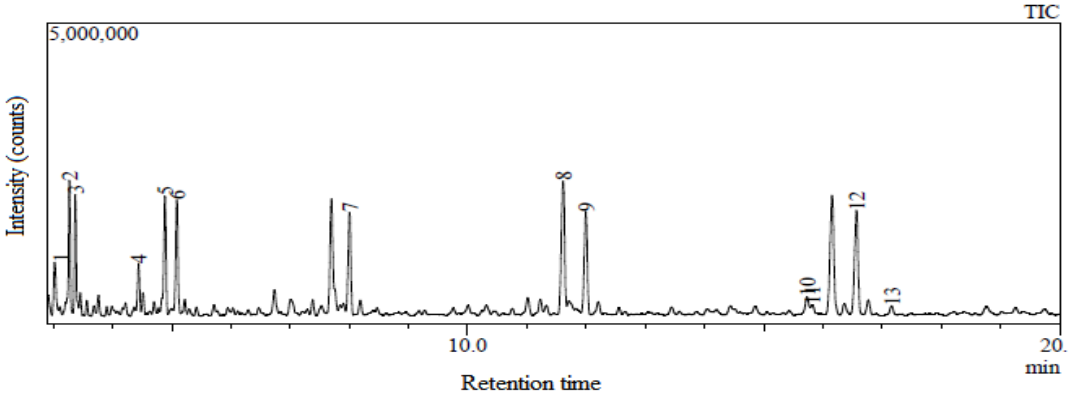

Figure 6. *Diagram chromatography of fuel oil from PE.*

(1): 2,4-Dimethylhexane, (2): 1-Heptene, (3): 2-Ethylpentane, (4): Toluene, (5): 1-Octen, (6): 2,4-Dimethylhexane, (7): Nonane, (8): 1-Decene, (9): Dodecane, (10): Cycloundecene (Z), (11): 2-Methyl-1-tetradecene, (12): Undecane, (13): 1-Undecene.

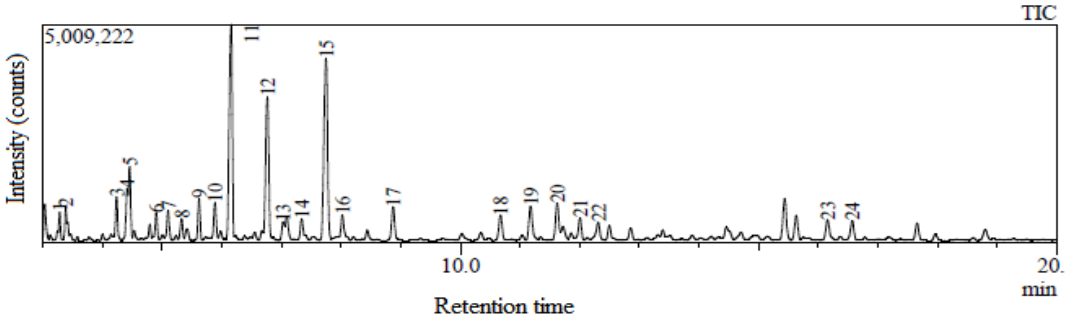

Figure 7. *Diagram chromatography of fuel oil from PP.*
(1): 1-Heptene, (2): 2-Ethylpentane, (3): 3,3-Dimethyl-1-hexene, (4): 2,3,4-Trimethyl-pentane, (5): Toluene, (6): 1-Octene, (7): 2,4-Dimethylhexane, (8): 3,3,5-Trimethyl-1-hexene, (9): 2,4-Dimethylundecane, (10): (2E)-4,4,5-Trimethyl-2-hexene, (11): (2E)-4,4,5-Trimethyl-2-hexene, (12): Ethylbenzene, (13): o-Xylene, (14): 5-Ethyl-2,4-dimethyl-2-heptene, (15): Styrene, (16): Nonane, (17): Isopropylbenzene, (18): Mesitylene, (19): alpha.-Methylstyrene,(20): 1-Ethyl-2-heptylcyclopropane, (21): 3,5-Dimethyloctane, (22): 2,5,5-Trimethylheptane, (23): 2,5,5-Trimethylheptane, (24): Dodecane.

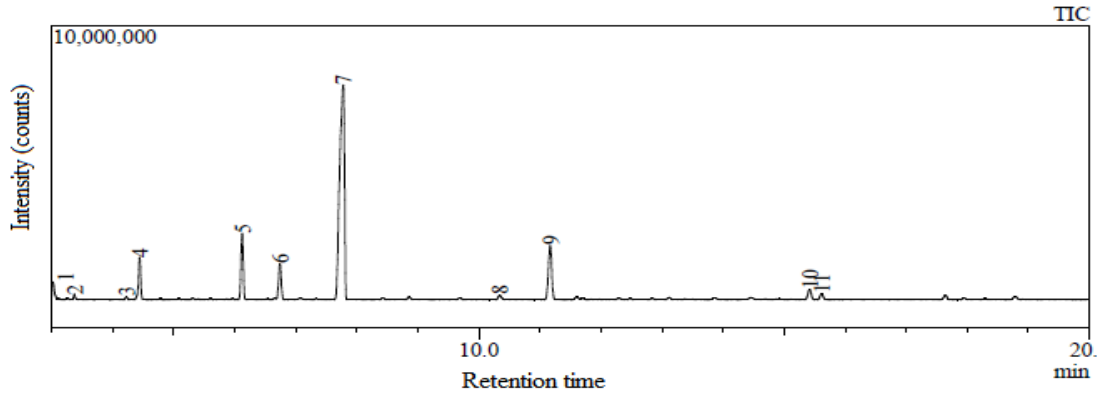

Figure 8. *Diagram chromatography of fuel oil from PS.*
(1): 2,4-Dimethylhexane, (2): Heptane, (3): (2E)-3,5-Dimethyl-2-hexene, (4): Toluene, (5): 2,4-Dimethyl-1-heptene, (6): Ethylbenzene, (7): Styrene, (8): Benzaldehyde, (9): alpha.-Methylstyrene, (10): 3-Octadecene, (11): 2,4-Dimethyl-1-decene.

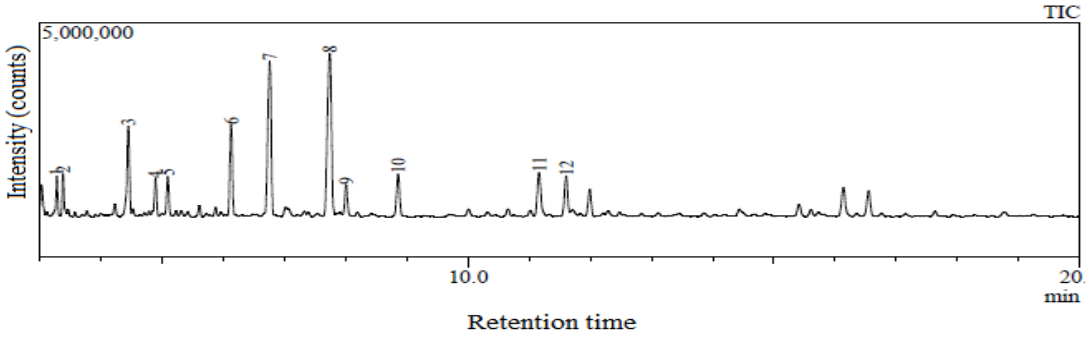

Figure 9. *Diagram chromatography of fuel oil from mixture plastic.*
(1): 1-Heptene, (2): Heptane, (3): Toluene, (4): 1-Octene, (5): 2,4-Dimethylhexane, (6): 2,4-Dimethyl-1-heptene, (7): Ethylbenzene, (8): Styrene, (9): Nonane, (10): Isopropylbenzene, (11): Isopropenylbenzene, (12): 1-Decene.

The result show that the majority of hydrocarbons in liquid oil were C7–C12, accounting for more than 90 area% of the total liquid oil. The composition of pyrolysis from PE, PP, PS and mixture plastics, primarily consisting of alkanes, alkenes, and aromatic compounds. In case of PS, the highest peak belong to styrene, indicating this is the main product of the pyrolysis process.

4. CONCLUSIONS

The study on the production of liquid gas from PP, PE, PS, and mixed plastic waste revealed that using a 500 W microwave power, a reaction time of 120 minutes, and a 10 wt% absorbent with a 20 wt% NiO/ZSM-5 catalyst, the resulting oil meets the specifications of FO No. 1 TCVN 6239:2019. Furthermore, the liquid fuel obtained has a efficiency of PS, PP reached 84%, PE reached 83%, and mixture plastics reached 83.5%. The result show that expanding the scale of pyrolysis and optimizing its efficiency and economic benefit are conductive to social development and environment improvement.

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TÓM TẮT

Chế tạo dầu nhiên liệu từ rác thải nhựa bằng nhiệt phân hỗ trợ vi sóng

Bài báo trình bày kết quả chế tạo dầu nhiên liệu từ rác thải nhựa bằng phương pháp nhiệt phân hỗ trợ vi sóng. Dưới ảnh hưởng của công suất, lượng chất hấp thụ vi sóng, thời gian nhiệt phân, xúc tác, rác thải nhựa polyetylen (PE), polypropylen (PP), polystyren (PS), hỗn hợp (34% PE +33% PP + 33% kl. PS) bị nhiệt phân thành dầu lỏng. Kết quả nghiên cứu cho thấy với 500 g nhựa nguyên liệu, công suất vi sóng 500 W, thời gian 120 phút, chất hấp thụ vi sóng cacbon chiếm 10% kl. (so với nhựa), xúc tác nhiệt phân chiếm 20% kl., hiệu suất chuyển hóa dầu nhiên liệu đạt 84% với nhựa PS và PP, đạt 83% với PE, 83,5% với nhựa hỗn hợp. Dầu thu được có các chỉ tiêu đạt tiêu chuẩn FO No1 TCVN 6239:2019. Kết quả phân tích sắc ký khí khối phổ (GC-MS) cho thấy thành phần của dầu nhiệt phân chủ yếu là hydrocacbon từ C7-C12.

Từ khoá: Rác thải nhựa; Nhiệt phân; Vi sóng; Dầu nhiên liệu.