Thermal processing of plastic wastes for fuel

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Abstract. Plastic wastes rise annually as a result of the growing demand for synthetic materials, which contributes to their manufacture. There are four main ways to recycle waste polymer, with thermal treatment for fuel being the most favorable to the environment. In this study, the thermal processing of plastic wastes was investigated with an anaerobic pyrolysis apparatus, and their thermal degradation was evaluated by using the thermogravimetric apparatus. Additionally, the elemental composition was determined by an elemental analyzer, n-alkanes were identified by gas chromatography with flame ionization detection/electron capture detector (GC-FID/ECD), and the hydrocarbons functional group was analyzed by Fourier transform infrared spectroscopy (FTIR). We pyrolyzed the most widely utilized polymers, including polypropylene (PP), low-density polyethylene (LDPE), and high-density polyethylene (HDPE), at temperatures as elevated as 500°C to obtain plastic pyrolysis oil (PPO). Then PPO was distilled into initial boiling point (IBP)-200°C (gasoline-like fuel), 200-350°C (diesel-like fuel), and over 350°C fraction (residue), and the technical features of each fraction were compared to the MNS 0217:2006 and MNS 6861:2020 standards. Diesel-like fuel (DLF) derived from LDPE consists of the n-alkane hydrocarbons with C8–C23 identified by flame ionization detection (FID) data; C10–C17 represented more than 80% of them. The hydrotreatment results revealed that the diesel-like fraction's nitrogen (N) and sulfur (S) amounts could have reduced from 0.06% to 0.01% and from 0.78% to 0.29%, respectively. In conclusion, it could be done to generate a product with a more stable hydrocarbon content from plastic wastes for fuel.

Keywords: Plastic wastes, thermal processing, plastic pyrolysis oil, gasoline, and diesel-like fuel, hydrogenation process, n-alkanes.

1   Introduction

A wide range of affordable, lightweight, and readily usable items that are necessary for daily life can be produced from plastic. The rate at which these products are manufactured has increased by nearly 9 percent annually [1].

The consumption of global plastic was estimated to be 430 million tons in 2019; statistical analysis indicates that this amount is expected to rise in the future, expanding

by 10 million tons per year to 460 million tons by 2023 [2]. Only 10% of these mass-produced material wastes are recycled, 14% are incinerated, and the rest end up in landfills and the environment [3], [4]. Widespread pollution from plastic in terrestrial and marine ecosystems is one of the world's most critical challenges because of inadequate management of plastic waste recycling [5], [6]. Plastic recycling processes fall into four categories: primary, secondary, tertiary, and quaternary. Above them, the third recycling method or tertiary category is thermochemical recycling, which is the most promising approach to waste treatment [7], [8], [9], [10].

Thermal processing of plastics primarily yields wax, solvents, oil, coke, or gases [11]. There are several benefits to using processed materials in fuel and gasoline operations, including the ability to recycle waste into useful products, increase oil reserves, and lessen the negative effects of plastic waste [12]. Recent studies have explored the possibility of obtaining fuel and gasoline by distilling the plastic pyrolysis oil [13], [14], [15]. Through the thermal process of plastics, various types of catalysts are used to improve the quantity and quality of gasoline and diesel-like fuel. For instance, researchers Ch. Kassargy et al. blended 50:50 ratio a of polypropylene (PP) and polyethylene (PE) with USY zeolite catalyst at a 10:1 ratio and pyrolyzed at 500 °C. Distilled gasoline-like fuel and diesel-like fuel obtained from blended plastic pyrolysis oil had a relatively high octane research octane number (RON = 89.8) and cetane number (45.1), respectively [16]. Likewise, V. L. Mangesh et al. pyrolyzed polypropylene with a metal catalyst (Ni-Mo) to produce a diesel-like fuel that matched EN590 standards (European diesel specifications) [17].

The quality of the obtained fuel depends mainly on the properties of the catalyst and raw materials. According to past studies, polypropylene pyrolysis oil has greater physicochemical characteristics than other polymer types. It is also easier to convert polypropylene pyrolysis oil into diesel fuel because of having a lower carbon chain number of the hydrocarbon compounds than other polymer types [18]. Plastic pyrolysis oil mostly contains alkene, alkane, and aromatic hydrocarbons [17]. Alkenes are highly reactive hydrocarbons that are unstable, which makes them an unsuitable component of diesel fuel [19]. To obtain the fuel more stable, the hydrotreatment process is required [20].

The process of hydrogen treatment of pyrolysis oil using sulfided $NiMo/Al_2O_3$ catalyst strongly depends on the reaction conditions, and if the temperature is too low (180°C), it leads to an increment of char yield [21]. Therefore, utilizing this catalyst for 2–6 hours at 300–350°C and 3–6 MPa of hydrogen pressure during the hydrotreatment process for PPO is more effective [17], [21], [22][23], [24]. The hydrogen processing method for improving the quality of raw materials is the most common method widely used in the processing of petroleum [25], biomass [26], coal [27], and other raw materials. However, the disadvantage of increasing the product's own cost after hydrogen treatment (hydrogen price 1.7–2.3 $/kg [28]) cannot be disregarded. So, the inclusion of each raw material in hydrogen processing is inefficient from an economic standpoint.

In this research, the most widely used polymer materials of polypropylene (PP), high-density polyethylene (HDPE), and low-density polyethylene (LDPE), were pyrolyzed to obtain PPO, which could be used to produce gasoline and diesel-like fuel. We focused on the study of changes in the composition of hydrocarbons and other

elements by hydroprocessing only the diesel fraction of LDPE with the presence of Ni-Mo/Al$_2$O$_3$ bi-metal catalyst, without including the PPO of each of the studied raw materials. The reasons behind this are as follows:

➢ Most of the characteristics of gasoline and diesel fractions obtained by pyrolysis of HDPE and PP were near the technical parameters of standards. Researchers Dobó Z, Kecsmár G, Nagy G, et al. revealed that gasoline-like fractions of PPO obtained by pyrolysis of LDPE HDPE and PP were near to the standard of the commercial fuel the (EN228) standard tests [24].

➢ Of the studied raw materials, only LDPE contained fuel contaminants such as nitrogen (N) and sulfur (S) in its products, hence its quantity had to be eliminated.

➢ In the work of the aforementioned researchers, only PPO was employed in the catalytic treatment. The hydrogenation process of diesel fraction recovered from polymers in the presence of Ni-Mo/Al$_2$O$_3$ catalysts has not been extensively studied and data on them lacking.

➢ Hydrotreating the target product would be economically feasible because hydrotreating PPO and all of its derived fractions has the drawback of increasing energy.

2   Experimental

2.1   Materials

A-HDPE   **B**-LDPE   **C**-PP

Figure 1. Materials of plastic wastes.

Materials of plastic wastes used in our research; LDPE; -LDPE plastic bags, HDPE; - bottles of milk, yogurt, juice, detergent, pills and vitamins, PP; -bags of cookies, chips, and chocolate, and disposable food containers.

2.2   Methods

Thermal processing (pyrolysis) experiments were performed at the pyrolysis apparatus shown in Fig. 2. A thermogravimetric instrument (Hitachi TG/DTA 7300) was used to determine the optimal conditions for the decomposition of waste plastics. The elemental composition of plastic wastes and diesel-like fractions obtained LDPE was determined by an elemental analyzer (FLASH™ 2000), and n-alkanes of diesel-like fractions were assessed by GC-FID/ECD gas chromatography (Agilent 7890A), and the functional groups of hydrocarbons were detected by FTIR (Alpha II, Bruker, Germany). The PPO was distilled into three fractions: gasoline-like fuel (IBP–200°C), diesel-like fuel (200–350°C), and residue (>350°C). The physical properties of each fraction were compared to MNS 6861:2020 and MNS 0217: 2006 (AI92) standards.
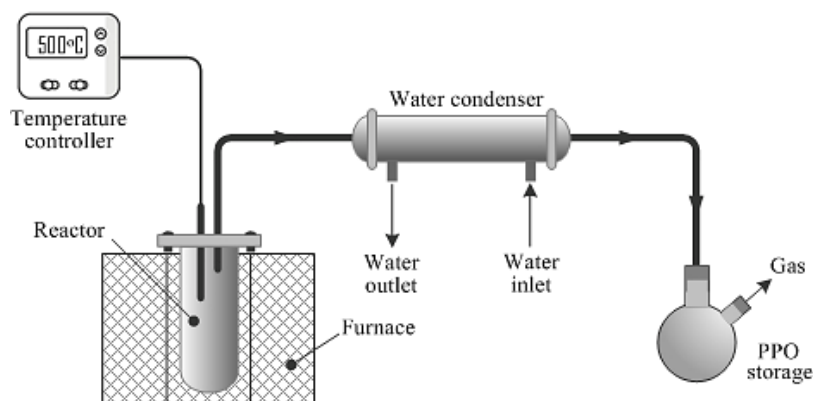

Figure 2. Pyrolysis apparatus [29].

2.3   Experimental procedures

2.3.1   **Sample preparation**: The collected waste was cleaned and washed. Then materials were dried in sunlight for 24 hours. After that, the samples were cut to size <3-5mm^2 and used for further thermal process.

2.3.2   **Pyrolysis experiment:** The scheme of the pyrolysis apparatus is illustrated in Fig. 2. About 100 grams of plastic waste were put into the pyrolysis reactor. Tighten the coupling to attach the straight cooler to the reactor. After setting the temperature of the apparatus, it can be ready for thermal processing. The reactor held two liters in capacity.

2.3.3   **Hydrotreatment process:** The hydrotreatment process was run with a catalyst and without a catalyst at 450°C for two and four hours under a hydrogen pressure of 5 MPa. The general scheme of the hydrotreatment process is shown in Fig. 3. A commercial Ni-Mo/Al$_2$O$_3$ catalyst (Ni 3%, Mo 15%) was used for the hydrotreatment process. About 5 g of diesel-like fuel obtained LDPE was charged into a batch-type micro-autoclave with an inner volume of 50 ml reactor, along with 1 wt.% Ni-Mo/Al$_2$O$_3$ catalyst and 1 wt.% sulfur.

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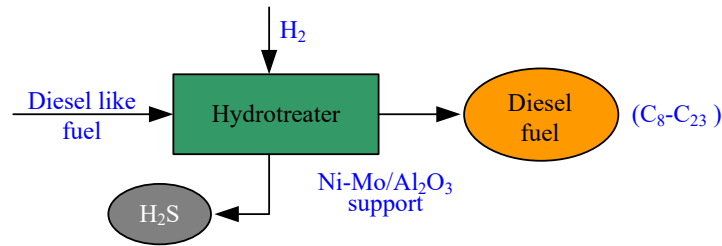

Figure 3. Scheme of hydrotreatment process

3 Experimental results

3.1. Results of ultimate and proximate analysis

Table 1 summarizes the results of the ultimate and proximate analysis of HDPE, LDPE, and PP.

Table 1. The ultimate and proximate analysis of plastic wastes.

Feedstock	Ultimate analysis (%)						Proximate analysis (%)			
	C	H	O	S	N	H/C	VM[*1]	A	FC[*2 a]	M[*3]
HDPE	85.03	14.14	0.83	0.00	0.00	1.99	99.28	0.72	-	-
LDPE	78.12	13.23	5.37	2.16	1.12	2.20	98.99	1.01	-	-
PP	84.36	15.02	0.62	0.00	0.00	2.14	99.51	0.49	-	-

[*1] Volatile matter, [*2] Fixed carbon, [*3] Moisture
[a]calculated by mass difference

Table 1 illustrates, the elemental composition of the waste plastics consists mainly of carbon atoms, and amounts of them decreased in the order of HDPE> PP>LDPE. The hydrogen amount of plastic waste was examined at 13.23-15.02, and the highest result was revealed for PP. The H/C ratio measures the proportion of paraffins, and naphthenes to characterize hydrocarbon fuels, and aromatics indicating their calorific value [30]. Results of elemental analysis revealed that oxygen amount was 0.62-5.37% for samples, but sulfur and nitrogen amounts were 2.16% and 1.12% for LDPE, respectively. Some researchers [31], and [32] revealed the amount of 11.25% hydrogen, 11.06% oxygen, 10.69% hydrogen, and 1.79% oxygen in the composition of LDPE, respectively. The research shows that the composition of elements varies depending on the purpose of raw materials. This is due to the amounts of sulfur, nitrogen, and oxygen included in various additives—which are used in diverse polymer production processes to improve plastic product quality and add color [33] [34] [35]. Proximate analysis revealed the content of volatile matter of plastic wastes was 98.99-99.51%. This indicates that the constituent parts of the polymer are almost entirely volatile

hydrocarbons (burnable), with a negligible ash content of 0.49-1.01% which can be explained by compounds containing non-volatile metals (Pb, Al, Ti, Zn, and others) added to its composition [36]. Thermochemical recycling is mentioned to be the most promising plastic recycling technique in the introduction; it involves thermally liquefying waste plastic in an anaerobic atmosphere to produce PPO that can be used as fuel in the future [7]–[10].

Thus, thermogravimetric analysis (TGA) analysis was carried out to determine the optimal conditions for the thermal decomposition of the plastic wastes, and the results are shown in the figure that follows.
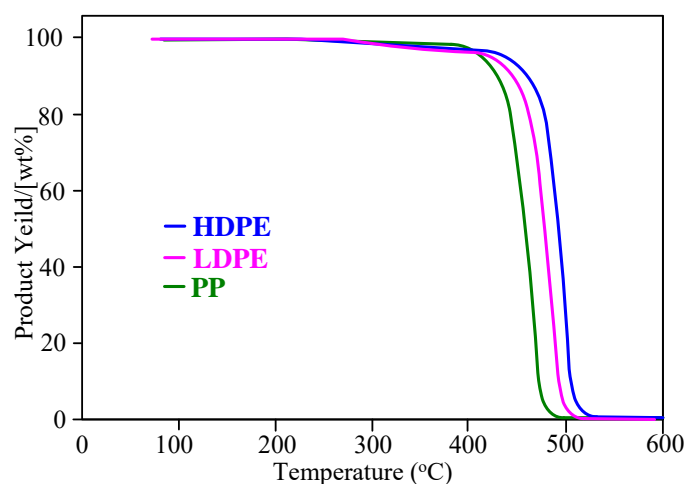

Figure 4. TGA analysis of plastic wastes.

According to Fig. 4, with degradation temperature occurring from around 220 °C, about 95% of HDPE decomposed at around 508 °C When the temperature increased up to 410°C, the first stage of LDPE degradation which began at about 280°C—achieved over 10% degradation. The second thermal degradation of LDPE began at around 400 °C, reached 50% degradation at 460 °C, and reached maximal decomposition (95%) at about 492°C (Fig. 4). PE is a long-chain material that degrades thermally via the random chain-breaking mechanism at comparatively higher temperatures [37]. Based on the PP thermal breakdown curve, approximately 95% of it underwent decomposition at 480°C, with the decomposition beginning at 210°C. Jung et al. [38] suggest that the tertiary carbons in the PP half-carbon chain enhance the carbocation process during thermal degradation, potentially causing it to degrade at a lower temperature. Additionally, according to Chandrasekaran et al. [39], differences in the TGA curves of different plastic feedstocks are caused by their structures and subsequent degradation mechanisms. Maximum degradation was shown at temperatures between 460°C and

500°C, as determined by TGA analysis of all the feedstocks. Numerous studies have conducted TGA analyses of plastic wastes [40], and our TGA results precisely correspond to their values. Section 3.2 provides a summary of the experiment findings regarding the yield and characteristics of plastic waste pyrolysis.

3.2. The yield of pyrolysis products and distilled fractions of PPO, and technical parameters of distilled fractions

Numerous studies are being conducted on the fuel processing of plastic waste, with the majority of these studies focused on the improvement of the liquid yield by plastic pyrolysis [41], [42], [43], [44]. The pyrolysis products of the waste polymers primarily consisted of PPOs (65.3-70.04%), while gas products were presented (29.24-34.05%). Fig.5 presents the results of the yield of pyrolysis products for plastic wastes.
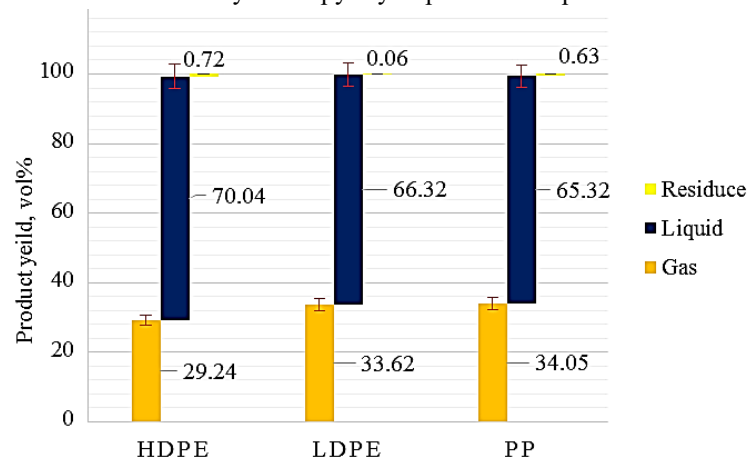

Figure 5. The yield of pyrolysis products for plastic wastes.

The yield of plastic pyrolysis products varies depending on the reactor type, its characteristics, and the desired output; some researchers could increase the PPO to 93%. [1]. However, there are relatively few research works that determine how the fuel produced by plastic processing is suitable for car engines. Researcher Bukkarapu et.al [45] investigated the possibility of using PPO obtained by plastic pyrolysis for diesel engines. Researchers found the following: obtained PPO has higher density and viscosity than commercial diesel fuel which lowers engine thermal efficiency, and emits more carbon monoxide when fuel burns. As a result, it cannot be used in the engine directly. However, when utilized in a blend of 80% PPO and 20% diesel fuel, it is suitable for the usage of diesel engines [45], [46]. However, direct mixing is inefficient because burning this blended fuel increased NO_x emissions by 25% and CO emissions by 5%. [46]. The reason for this is explained by the researcher Bukkarapu et

al [45] due to the high ratio of carbon to hydrogen (H/C) in the combustion fuel in the internal combustion engine system and the low oxygen supply, resulting in a long ignition time of the fuel [47].

We distilled PPOs into three fractions gasoline-like fuel (IBP-200^{0}C fraction), diesel-like fuel (200-350^{0}C fraction), and the residue (above 350^{0}C fraction), and the yield of fractions is shown in Fig. 6.
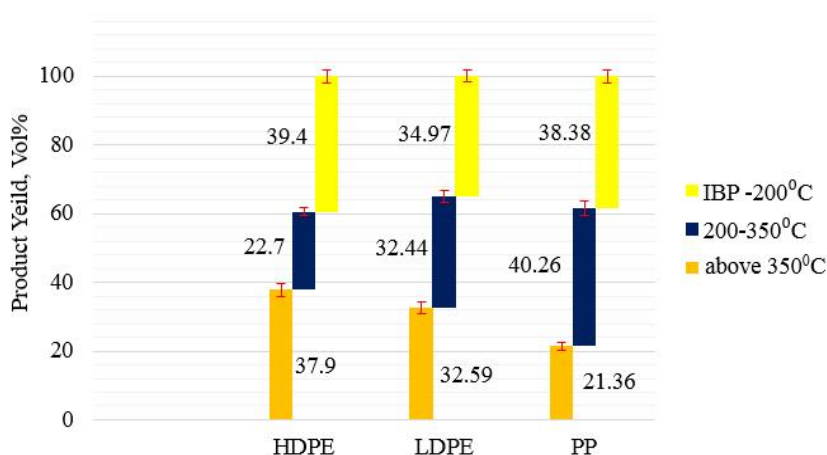

Figure 6. The yield of distilled fractions of PPOs obtained by plastic wastes.

We can see that the yield of gasoline-like fuel was nearly similar about 34.97-39.4% for different feedstocks, the yield of the diesel fraction was the highest for PP (40.26%), and the lowest result was for HDPE 22.7%. The yield of residue fraction decreased in the sequence of HDPE>LDPE>PP. From the yield of the fractions obtained from the PPO of HDPE, it can be seen that the content of gasoline fraction and residual fraction occupies the majority (77.3%), which reveals more suitable for processing gasoline and furnace fuel. Researchers J. M. Escola et al. found that the diesel fraction had a yield of HDPE-34%, LDPE-32%, and PP-28, and the gasoline fraction had a yield of HDPE-39%, LDPE-44%, and PP-58% [47].

Table 2 presents the technical parameters of the gasoline-like fuel of PPOs derived from different plastic wastes.

As shown in Table 2, octane numbers of gasoline-like fuel were 84-89. Some researchers determined that the octane numbers of gasoline-like fuel obtained by PPOs were HDPE-80.6, LDPE-80.5, and PP-93.4 respectively [48]. Also, researcher Kassargy C et al. found out the octane number of the gasoline fraction obtained by pyrolysis of PP and PE-1:1 was 89.9 [49].

Table 2. Technical parameters of the gasoline-like fuel or IBP- 200°C fraction obtained by different plastic wastes.

Technical parameters	Units	MNS 0217 : 2006 (RON92)	IBP-200^{0}C fraction of;		
			HDPE	LDPE	PP
Research octane number, (RON)	-	92 (min)	86	84	89
Motor octane number, (MON)	-	83 (min)	82	72	78
Density @ 15°C	g/cm^3	725-780	782	740	730
Distillation data (summer)					
- initial boiling point	°C	35 (min)	58	38	32
- distillation temperature T_{10}	°C	75 (max)	95	78	73
- distillation temperature T_{50}	°C	120 (max)	134	123	108
- distillation temperature T_{90}	°C	190 (max)	194	182	175
Final boiling point (FBP)	°C	215 (max)	221	218	210
Distillation residue	% vol.	2 (max)	2.2	2.8	2.1

Their outcomes were quite similar to our results. The gasoline fraction obtained by thermal processing of polypropylene (PP) has a relatively high octane number [50]. The high octane number of gasoline-like fuel of polypropylene reflects its branched structure. The prevailing hydrocarbon (58.4 mass %) is 2,4-dimethyl-1-heptene for the PP distillate [51]. Mainly 1-alkenes are carriers of the octane level in the case of naphtha fractions obtained from polyethylenes [48]. The density of the gasoline fraction obtained from the investigated plastics was increased in the order PP<LDPE<HDPE and their value ranged from 730-782g/cm^3. It can be seen the densities of the gasoline-like fuel were within the limits of MNS 0217: 2006 standard for PP and LDPE. This can be explained that there are lower hydrocarbon chains in the compounds generated during the decomposition of PP and LDPE than in HDPE.

We also determined the technical parameters of the diesel-like fuel of different PPOs, and compared it with commercial diesel fuel standard (MNS 6861:2020). The results are summarized in Table 3.

Table 3. Technical parameters of the diesel-like fuel or 200-350°C fraction obtained by PPOs of different plastic wastes.

Technical parameters	Units	MNS 6861:2020 (winter)	200-350^{0}C fraction of;		
			HDPE	LDPE	PP
Cetane number	-	45	45	43	42
Distillation data					
Distillation temperature T_{50}, max	°C	280(max)	281	288	278
Distillation temperature T_{96}, max	°C	340(max)	344	355	346
Density @25°C	kg/m^3	860	820	814	802
Viscosity @ 20 °C	cCт	3-6	4.09	5.85	5.66

Table 3 shows that the cetane numbers of the diesel-like fuel or 200-350°C obtained by distillation of plastic wastes were 42-45 which of them was near to the MNS standard specification. Combustion properties of diesel fuel are described by the cetane [48]. Some researchers found that diesel fractions produced by the processing of plastics generally have high cetane numbers like 51.6 and 84 [52]. The high cetane number is represented by low aromatic content in fuel [53]. As can be seen in Fig. 7, the absorption of functional groups of chemical components in the composition of distilled fractions obtained by PP feedstock by FTIR.
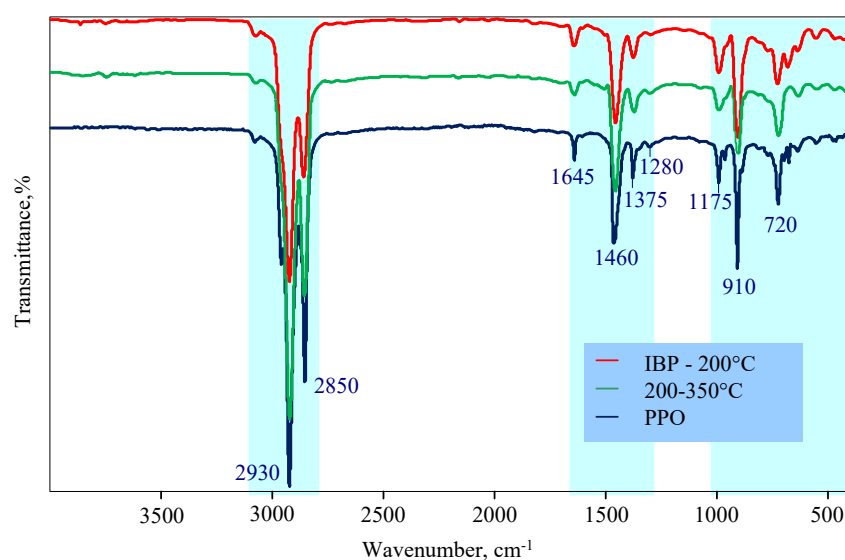

Figure 7. FT-IR analysis of polypropylene

Fig.7 shows the compared FT-IR results of PPO, gasoline-like fuel, and diesel-like fuel. The strong peaks at 2930 cm^{-1} and 2850 cm^{-1} were attributed to the presence of methylene groups (CH_2) [54]. The peak at 1460 cm^{-1} was attributed to the asymmetric deformation vibrations of CH_2 and CH_3 groups. In addition, the peak at 1375 cm^{-1} was attributed to the bending vibration of CH_3 groups [55]. The weak peak located at 1645 cm^{-1} indicated the presence of alkene [54]. Many papers confirmed the strong peak at 720 cm^{-1} was attributed to aromatic stretching vibration [56], [57]. The vibration located at 910 cm^{-1} was attributed to the di-substituted benzene. From FTIR results, it was confirmed that the pyrolysis products of waste plastic were the main alkane and arenes.

Results of elemental analysis revealed LDPE contains sulfur (2.16%) and nitrogen (1.12%) atoms. Section 3.3 provided further details on the process of catalytic hydrotreatment process of the diesel-like fuel obtained PPO of LDPE to eliminate the components of sulfur and nitrogen.

3.3. Hydrotreatment process of diesel-like fuel derived from LDPE

Diesel-like fuel derived from LDPE was further hydrogenated for two and four hours in the presence of Ni–Mo/Al$_2$O$_3$ (Ni 3%, Mo 15%) catalyst. Sulfur and nitrogen heteroatoms must be removed from the fuel's composition to obtain high-quality fuel [58]. The following Figures illustrate the results of the hydrotreatment process with the catalyst. We can see the appearance of fractions, and changes in the distribution of n-alkane hydrocarbons in Fig. 8 and, 9 respectively.
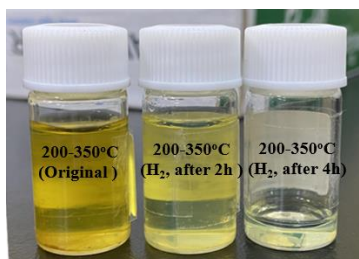

Figure 8. The appearance of fractions
(results of hydrotreatment process with the catalyst).

As shown in Fig. 8, it was evident that the color of DLF derived from LDPE varied significantly after the hydrotreatment process.
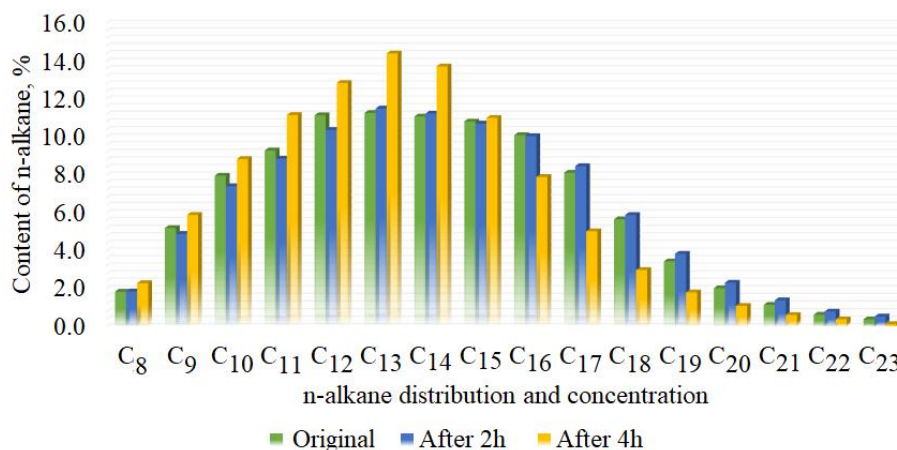

Figure 9. The distribution of n-alkane hydrocarbons of diesel-like fuel derived from LDPE (results of hydrotreatment process with the catalyst).

Diesel-like fuel obtained by pyrolysis of LDPE detected hydrocarbons with carbons ranging from C$_8$ to C$_{23}$. The majority (almost 80%) of the n-alkane hydrocarbons detected were of C$_{10}$–C$_{17}$ hydrocarbons. After hydrotreatment with Ni-Mo/Al$_2$O$_3$ for

four hours, the amount of C_8–C_{15} carbon increased from 68.5% to 80%, but the distribution of C_{16}–C_{23} decreased by 11.5%. According to the study of Bayanmunkh M, et al., petroleum atmospheric residue (>350°C) was hydrotreated using a catalyst of Ni-Mo/Al2O3, and the yield IBT-350° was increased from 33.5% to 38% [59]. The majority of the composition of diesel fuel is occupied by aliphatic hydrocarbons with $C_{10}H_{20}$-$C_{15}H_{28}$, their content is about 75%, while the remaining 25% consists of styrene and benzene derivatives [60]. Therefore our findings indicate that the composition of diesel fraction hydrotreated was improved nearly to the composition of standard diesel fuel.

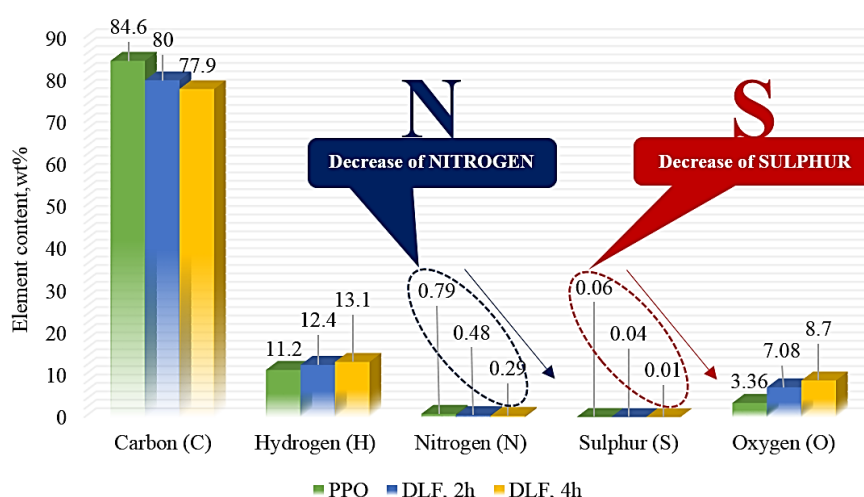Fig.9 illustrates comparable results of elemental analysis for DLFs.

Figure 10. Results of elemental analysis for DLFs.

After the hydrotreatment process in the presence of Ni-Mo/Al$_2$O$_3$ catalyst for 2-4 hours, the elemental composition of PPO and DLF is shown in Fig. 10. It can be seen the carbon amount decreased from 84.6% to 77.9%, but hydrogen (H) atoms increased from 11.2% to 13.1% for PPO and DLF (4h), respectively. The sulfur content was defined as 0.2% in commercial fuel under the MNS 6861:2020 standard requirement. The sulfur (S) content of the diesel fraction obtained by pyrolysis of LDPE was 0.06%, which could be reduced to 0.01% after the catalyst hydrotreatment process. Also, the amount of nitrogen (N) atoms decreased from 0.78% to 0.29%. The inclusion of diesel fraction in the hydrotreatment process has many advantages, such as the removal of heteroatoms of sulfur and nitrogen in its composition, and the conversion of unsaturated hydrocarbons with high chemical activity into saturated hydrocarbons to make the composition of the fraction more stable.

4 Conclusion

In this study, we pyrolyzed the most widely utilized polymers, including PP, LDPE, and HDPE, at temperatures as elevated as 500°C to obtain plastic pyrolysis oil (PPO). Then PPO was distilled into IBP-200°C (gasoline-like fuel), 200-350°C (diesel-like fuel), and fractions over 350°C, and the technical parameters of each fraction were compared to the MNS 0217:2006 and MNS 6861:2020 standards. DLF derived from LDPE consists of the n-alkane hydrocarbons with C_8–C_{23} identified by FID data; C_{10}–C_{17} represented more than 80% of them. The hydrogenation results for four hours indicated that the diesel-like fraction's nitrogen (N) and sulfur (S) amounts could have reduced from 0.06% to 0.01% and from 0.78% to 0.29%, respectively. In conclusion, it could be done to generate a product with a more stable hydrocarbon content from plastic wastes for fuel.

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