



Thermal Pyrolysis of Waste Polyolefins to Generate Alternative Liquid Fuel

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Abstract

Waste plastics are the cause of great environmental issues because they are non-biodegradable. In this work, waste polyolefins plastics of high-density polyethylene (HDPE), low-density polyethylene (LDPE), and Polypropylene (PP) were separately subjected to pyrolysis in a fixed bed batch reactor within the temperature range of approximately 300 – 510 °C and heating rate of 12 °C/min. The result of the pyrolytic oil obtained was characterized and all yields above 50 % in terms of the liquid oil. PP with 58.6 % has the highest oil yields slightly above LDPE with 54.3 % and HDPE with 51.5 %. The gaseous effluents calculated by the difference between the oil collected and char residue were higher for HDPE at 43.4 %, followed by LDPE at 39.2 % and PP at 36.8 %. The analysis of pyrolytic oil and Liquid fuel shows that the specific gravity, pour point, and flashpoint are 0.775, -4.5 °C, 23 °C respectively. Specific gravities were slightly lower than the lower limit of the Department of petroleum resources (DPR) approved a range of 0.820 but approximately equal to that of virgin gasoline. However, the kinematic viscosity @ 40 (mm²/sec) and the flashpoints were within the approved range. The investigation revealed that the GC-MS analysis of the oils to contain aliphatic and cyclic hydrocarbons in the range of Diesel and kerosene fuel. Hence waste plastic can serve as an alternative source of energy.

Keywords

Thermal pyrolysis; alternative energy; batch reactor; pyrolytic oil; plastic wastes; polyolefin.

1.0 Introduction

Plastics have grown into an indispensable and crucial material in this present age because of the vital role it plays in enhancing human way of life both in industrial, manufacturing, commercial and residential sector such as in packaging materials, agriculture, healthcare, building, lagging, and insulation, automobile sector, textiles, sports equipment, toys, and electronics devices (Association of Plastic Manufacturers Europe, 2015; Michael, 2015). Also, due

to its unique material properties, plastics are employed in making products that are lightweight, toughness, water-resistant, chemical inert, temperature resistant, and having-low electrical and thermal conductivity (Fakhrhoseini and Dastanian, 2013; Shafferina *et al.*, 2017). Based on plastics behaviour when heated, plastics are divided into two main categories: thermoplastic and thermosetting plastics (Lopez *et al.*, 2011 British Plastics Federation, 2015). Polyolefins (POs) which include high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP) are commonly used for many everyday plastic commodities. Most of these plastic products normally result in the waste stream just after a single use which makes POs significant in plastic waste. Therefore, the number of plastics and other polymeric materials collected in the environment every year is of frightening proportion (Williams and Williams, 2009; Michael, 2015; Bridwater, 2012; Fakhrhoseini and Dastanian, 2013). This makes waste plastic a good potential feedstock for alternate diesel and gasoline production (Abbas-Abadi and Mcdonald, 2014; Dayana *et al.*, 2016).

Pyrolysis is referred to as the thermal degradation of biomass which requires a little duration of intense heat (at elevated temperature and low pressure) in the absence of oxygen (Fakhrhoseini and Dastanian, 2013; Shafferina *et al.*, 2016; Stelmachowski, 2016). The process drastically reduces the initial quantity of the waste and yields other energy carriers in form of pyrolysis oil, carbon black, and hydrocarbon gases (Cleetus *et al.*, 2013; Shafferina *et al.*, 2017) which are beneficial for industries mostly production and refineries. Most plastic pyrolysis in the laboratory scale was performed in batch or semi-batch reactors (William and William, 2009; Shafferina *et al.*, 2017). Pyrolysis in the batch reactor is normally performed at a temperature range of 300 – 800 °C for both thermal and catalyst pyrolysis (Jan *et al.*, 2010; Shah *et al.*, 2010).

Catalysts are important in the catalysis pyrolysis of plastics (Kayacan and Dogan, 2008; Onwudili *et al.*, 2009; Jan *et al.*, 2010) as they reduce the activation energy of the process, thus expedite the rate of reaction and improve the yield of the pyrolysis process (Adai, 2010; Stelmachowski, 2016). The usage of catalysts for thermal pyrolysis plastics in a batch reactor as used in this research work is unnecessary because there would be a high tendency of coke formation on the surface of the catalyst which reduces the catalyst efficiency overtime and also caused high residue in the process. Also, there will be a challenge to separate the residue from the catalyst at the end of the experiment (Fakhrhoseini and Dastanian, 2013; Shafferina *et al.*, 2017). Several scholars

have worked extensively on pyrolysis plastics temperature (Uddin *et al.*, 1996; Miranda *et al.*, 2001; Scheir and Kaminsky, 2006; Othman *et al.*, 2008; Marcilla *et al.*, 2009; Jan *et al.*, 2010; Jung *et al.*, 2010; Shah *et al.*, 2010; Branch and Azad, 2013; Abbas-Abadi *et al.*, 2014; Shafferina *et al.*, 2017). Kumar and Singh, (2011) have also studied the HDPE thermal pyrolysis at a higher temperature range of 400-550 °C. These researchers' findings proved that pyrolysis liquid oil could be obtained at a higher temperature beyond 510 °C mostly for the thermal pyrolysis method. Mastral *et al.*, (2001) also conducted pyrolysis of PVC on a batch reactor at a temperature range of 225-520 °C. Thermal degradation behaviours of the plastics can be measured using thermogravimetry analyzer (DTC) curves based on derivative thermogravimetry (Shafferina *et al.*, 2017).

The amount of plastic wastes available in every country is reaching millions of tons (Shafferina *et al.*, 2017). Globally, the total plastic production has increased by nearly 200 times from 2 million tonnes per year in 1950 to 381 million tonnes per year in 2015, the demand for plastic is predicted to double in the next 20 years as the world population rises (Lebreton and Andrady, 2019; Rahman *et al.*, 2019; Hannah and Max, 2020). This huge appetite for plastic has resulted in plastic waste pollution with non-recyclable plastic leaking at an unprecedented rate into the groundwater, land, and marine environment as well as in deep remote oceans such as the Marianas Trench. Mitigating actions such as the ban on single-use plastic, using biodegradable plastics, and improved waste management are often faced with business, civil, or government resistance challenges (Williams and Williams, 2009; Michael, 2015; Delvin, 2011; Linda, 2019). For instance, in Nigeria, the quantity of solid wastes generated annually is put at 32 million tonnes, representing one of the highest in Africa, and only 9 % is being recycled (Rahman *et al.*, 2019). Of this figure, plastic constitutes 2.5 million tonnes, much of which will inevitably end up in the waste streams. Hence, waste plastics are endemic environmental concerns. Therefore, the thermal pyrolysis in a locally fabricated pyrolysis unit on different types of waste POs plastics such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP) to produce liquid oils suitable for diesel oil will concomitantly reduce the environmental effect of the plastic wastes.

2.0 Materials and Methods

2.1 Samples collection

The samples collected for the experiment are components of the polyolefins: Sachet water films were collected as a sample of low-density polyethylene (LDPE), while different plastic containers were collected for high-density polyethylene (HDPE) and polypropylene (PP), with clear respective types labelled. They were all obtained from the post-consumer waste stream.

2.2 Description of the pyrolysis reactor

A locally fabricated fixed bed batch reactor with external heating was used for the pyrolysis process to produce oil. It is a simple laboratory-scale reactor made up of an extended stainless-steel pipe of length (11.5 inches) and diameter (14 inches) respectively. The designed reactor was covered at the top to condense the liquid product for collection, a small safety relief valve passage was attached to the lid of the reactor for the release of the gas. A pipe was connected to the chamber via a flange joint created by a rim at the end of the pipe. Other basic components used for the construction of the pyrolysis chamber included a stainless-steel fixed bed, temperature controller, condenser, temperature sensor, gas heater, insulator, storage tank, valve, gas exit line. The thermocouple temperature sensor was used inside the pyrolysis chamber to monitor the temperature when heated by an external source. The presence of oxygen will make the plastic samples burn rather than melt; hence, the reactor was well lagged and leakages free. Liquefied petroleum gas (LPG) was used as a heating medium for the process. As there is an increased in temperature, the plastics melted, and the vapour rises gradually. Through the steel pipe, the gas condensed to liquid oil which is gently collected into the beaker.

2.3 Experimental procedure

The three sets of samples were treated one after the other. The plastic materials were sorted, shredded into pieces by cutting using a knife, washed to remove foreign materials like oil and mud, and drying of the moisture in the open air. Samples of 1 kg were fed into the batch reactor at each experimental cycle. The reactor was tightly covered with aid of bolt and nuts using a spanner, to prevent the flow of air, an external heat was then applied by putting the reactor on the LPG fired stove. The temperature gradually increased until about 310 °C, when drops of condensed oil were first noticed. The process was

allowed to continue for 45 minutes while the Liebig condenser was connected to the reactor to condense the vapour escaping the combustion chamber (oil) into a container. While the heating continued, the maximum temperature was reached at 510 °C and a heating rate of 12 °C/min. The fire was then put off, and the reactor was left to cool down before opening. At first, the residues were a viscous liquid which solidified into wax after further cooling. The mass of the oil collected, and the char residue were directly determined by weighing, while that of gas produced was found by using Equation (1.0).

$$\text{Mass of gas (g)} = [\text{Mass of sample (g)}] - [\text{Mass of Oil (g)}] - [\text{Mass of Tar (g)}] \quad 1$$

The gaseous effluents calculated by differences were higher for HDPE at 43.4 % followed by LDPE at 39.2 % and PP at 36.8 %, These may be attributed to the nature of the chain length of the individual plastics. The percentage yield in which oil is considered the desirable product was obtained by Equation (2.0).

$$\text{Yield (\%)} = \text{product (g)} / \text{Feed (g)} \times 100 \quad 2$$

The weight of collected oil and the recovered char (residue) from each of the waste material treated were weighed and the results collected. Figure 1 shows the scheme of the process involved in the experimental setup.

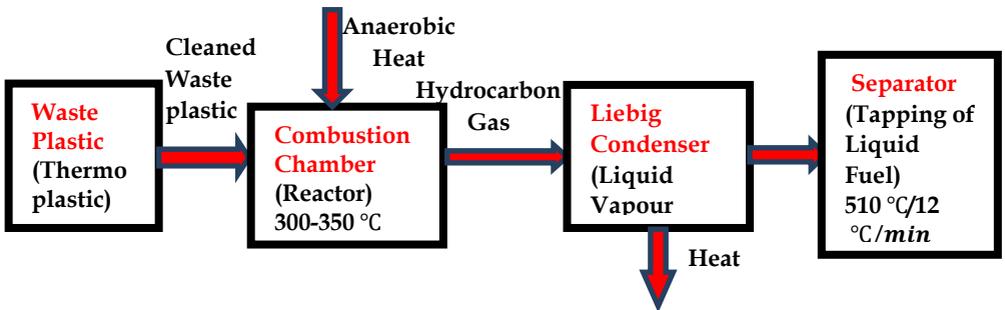


Figure 1: Simplified flowchart of non-biodegradable waste plastic conversion into fuel by pyrolysis process.

3.0 Results and Discussion

3.1 Proximate and ultimate analyses of plastic used

The waste plastics were washed and shielded before the ultimate and proximate contents were determined using the methods described by Kreith, 1998; Othman *et al.*, 2008 and Jung *et al.*, 2010 to measure the chemical properties and ascertain the elements of the plastic compound of the HDPE, LDPE, and PP waste materials. The ultimate and proximate analysis of polyolefin wastes contains mainly carbon and hydrogen as shown in Table 1. The proximate analysis reveals that the volatile matter and ash content greatly affect the liquid oil production through the pyrolysis process; while ash content lessens the liquid oil produced, the volatile matter increases the oil yield (Regnier *et al.*, 1995; Shafferina *et al.*, 2016).

Table 1: Ultimate analyses of different plastic materials

Type	Mark	Ultimate						Proximate			
		C	H	N	S	O	Cl	Moisture	Fixed-carbon	Volatiles	Ash
HDPE		88.40	10.72	0.18	0.04	0.73	0.02	0.02	0.06	98.82	1.12
LDPE		83.96	14.47	0.29	0.18	0.71	0.02	0.03	0.08	98.49	1.40
PP		84.71	14.42	0.25	0.02	0.28	0.01	0.03	0.12	98.66	1.60

3.2 Process yields of the pyrolysis

Heat is supplied above the melting point of each polyolefin material used in the absence of air, resulting in random breakages of the polymer chains. Within the range of the temperatures in which the pyrolysis was carried out, the percentage of oil yields was above 50 % for all three types of polymeric waste pyrolysis. The material tested is good for producing oil. PP has slightly highest oil yields of the three materials investigated. This is expected, and probably because the entire chain length of PP is completely saturated with single bonds. This may be the reason for its lowest char yield. The yield of the oil and other products of the pyrolysis are presented in Table 2. The gaseous by-product of the pyrolysis process has economic and appreciable calorific

values which can be reused to improve the overall energy requirement of the pyrolysis industry plant, used in gas turbines to generate electricity and direct firing in boilers without need for flue gas treatment (Fernandez *et al.*, 2011; Branch and Azad, 2013).

Table 2: Percentage yield of the pyrolysis

Material	Oil (%)	Char (%)	Gases (%)
HDPE	51.5	5.1	43.4
LDPE	54.3	6.5	39.2
PP	58.6	4.6	36.8

The obtained temperature of 510 °C of this study was further proven by Demirbas (2004) who carried out the PP pyrolysis at extreme temperatures of 740 °C in a batch reactor which resulted in 48.8 % wt. liquid yield, 49.6 %wt. of gaseous and 1.6 %wt. char. However, the energy balance for a laboratory-scale approach such as obtainable in this research is unnecessary but it is important for the design of larger unit applications including specifications for coolers and quench units as stated by Atsonios *et al.*, 2015.

3.3 Characterization of the Pyrolysis Oil

The oil from each of the products was subjected to an analytical test of major parameters and to ascertain whether each batch of product produced conform to the specification, suitable for marketing purposes. It also helps to determine if there was any contamination on the produced oil that will harm the quality of the products. The characterization of the pyrolysis oil samples from each plastic material was subjected to standard laboratory tests to determine the specific gravity, flashpoint, viscosity index, and pH values. This was carried out at the NNPC Mosimi Depot laboratory. Viscosity was determined by a Rotational viscometer equipped with an SC4-18 spindle (Brookfield Viscometer model DVII + Pro EXTRA) at 40 °C. Density was obtained using a 25 ml pycnometer and the pH of the samples was measured using Mettler Toledo pH meter series 320. From the comparative analysis, it is clear that the properties of pyrolysis fuel are similar to that of diesel. Hence plastic pyrolysis fuel can be used as an alternative fuel for diesel engines and oil-fired furnaces. Table 3 shows pyrolytic oil parameters compared to the standards.

Table 3: Pyrolysis oil analytical tests

Parameter	Unit	HDPE	LDPE	PP	Pyrolytic oil	Diesel	Petrol	AGO
Specific gravity		0.8020	0.7650	0.810	0.775	0.82	0.775	0.820 - 0.870
Density	(Kg /m ³)	968	942	853	775	820	770	800
K.V @40	mm ² /sec	0.33	0.53	0.27	0.48	2.2	0.55	1.6 - 5.5
Flash Point	(° C)	10	24	40	23	72	-43	66
Pour point	(° C)	17.8	15	13.5	-4.5	-4.6	-4.6	6
pH		6.8	6.5	6.6	-	-	-	-

Note: K.V – Kinematic viscosity

3.4 HDPE, LDPE, and PP GC-MS Spectral analysis

The pyrolysis oil samples were analyzed in Agilent 7820A GC-MS with column HP-5ms ultra inert (30 m × 250 μm × 0.25 μm) using 99.9 % Helium carrier gas at 19.604 psi at a flow of 2.5 ml/min. The oven temperature was held at 40 °C for 4 mins and ramped up at 15 °C/min to 250 °C for a holding time of 10 mins. The total run time for each sample was 28 mins. The compounds in the chromatogram generated were identified with the NIST database and peaks with a minimum of 90 % probability were only considered in this study. The results of the GC-MS analysis of the HDPE, LDPE, and PP pyrolysis oil samples indicated 468, 518, and 522 peaks respectively. However, 72, 81, and 71 peaks were with a minimum of 90 % quality. The result of the HDPE pyrolysis oil contains aliphatic and cyclic hydrocarbons, esters, haloalkanes, esters, alkenone, alkanals, and other complex organic compounds, the hydrocarbon is in the range C₁₀ – C₃₆ and consists of majorly diesel, kerosene, and heavy oil. This result is in agreement with the previous study on the pyrolysis of HDPE (Kumar and Singh, 2011). While the LDPE pyrolysis oil was found to contain aliphatic hydrocarbons, halogenated alkanes, and other compounds, the hydrocarbons were observed to range between C₁₂ – C₃₈ with most of the compounds in the kerosene and diesel range. Most of the compositions were in agreement with previous studies on LDPE pyrolysis oil analysis (Sarker *et al.*, 2012 and Eletta *et al.*, 2017). The hydrocarbon in PP pyrolysis oil contains aliphatic hydrocarbons, cyclic hydrocarbons, acyclic alkenes, haloalkanes, and other complex compounds. The PP oil yield majorly diesel fractions, gasoline fractions, kerosene fractions with little heavy oil compared to HDPE and LDPE as also reported in previous studies (Shafferina *et al.*, 2016; Stelmachowski,

2016). Table 4 summarized the key properties of the fuel oil using the GC-MS analysis of the studied plastics.

Table 4: The key constituents of GC-MS results of the pyrolysis oil

HDPE		LDPE		PP	
Compound	Formula	Compound	Formula	Compound	Formula
13-Methylheptatriacontanane	C ₃₅ H ₇₀	13-Methylheptatriacontanane	C ₃₈ H ₇₈	13-Methylheptatriacontanane	C ₃₈ H ₇₈
13-Methyl-Z-14-nonacosene	C ₃₀ H ₆₀	17-Pentatriacontene	C ₃₅ H ₇₀	17-Pentatriacontene	C ₃₅ H ₇₀
1-Decene	C ₁₀ H ₂₀	1-Docosene	C ₂₂ H ₄₄	1-Decene	C ₁₀ H ₂₀
1-Docosene	C ₂₂ H ₄₄	1-Eicosene	C ₂₀ H ₄₀	1-Docosene	C ₂₂ H ₄₄
1-Dodecene	C ₁₂ H ₂₄	1-Heptadecene	C ₁₇ H ₃₄	1-Hexacosene	C ₂₆ H ₅₂
1-Eicosene	C ₂₀ H ₄₀	1-Hexacosene	C ₂₆ H ₅₂	1-Nonadecene	C ₁₉ H ₃₈
1-Heptadecene	C ₁₇ H ₃₄	1-Nonadecene	C ₁₉ H ₃₈	1-Octadecene	C ₁₈ H ₃₆
1-Hexacosene	C ₂₆ H ₅₂	1-Octadecene	C ₁₈ H ₃₆	1-Tetracosene	C ₂₄ H ₄₈
1-Nonadecene	C ₁₉ H ₃₈	1-Pentadecene	C ₁₅ H ₃₀	1-Tridecanethiol	C ₁₃ H ₂₈ S
1-Octadecene	C ₁₈ H ₃₆	1-Tetradecene	C ₁₄ H ₂₈	1-Tridecene	C ₁₃ H ₂₆
1-Pentadecene	C ₁₅ H ₃₀	1-Tricosene	C ₂₃ H ₄₆	2-Dodecene, (Z)-	C ₁₂ H ₂₄
1-Tetracosene	C ₂₄ H ₄₈	2-Methyl-Z-4-tetradecene	C ₁₅ H ₃₀	2-Tetradecene, (E)-	C ₁₄ H ₂₈
1-Tridecene	C ₁₃ H ₂₆	2-Tetradecene, (E)-	C ₁₄ H ₂₈	3-Eicosene, (E)-	C ₂₀ H ₄₀
3-Heptadecene, (Z)-	C ₁₇ H ₃₄	3-Eicosene, (E)-	C ₂₀ H ₄₀	5-Octadecene, (E)-	C ₁₈ H ₃₆
8-Heptadecene	C ₁₇ H ₃₄	3-Heptadecene, (Z)-	C ₁₇ H ₃₄	5-Undecene	C ₁₁ H ₂₂
Cyclodecane	C ₁₀ H ₂₀	4-Tetradecene, (E)-	C ₁₄ H ₂₈	6-Tridecene, (Z)-	C ₁₃ H ₂₆
Cyclododecane	C ₁₂ H ₂₄	6-Tridecene	C ₁₃ H ₂₆	8-Heptadecene	C ₁₇ H ₃₄
Cyclohexadecane	C ₁₆ H ₃₂	7-Hexadecene, (Z)-	C ₁₆ H ₃₂	Cyclodecane	C ₁₀ H ₂₀
Cyclooctacosane	C ₂₈ H ₅₆	8-Heptadecene	C ₁₇ H ₃₄	Cyclododecane	C ₁₂ H ₂₄
Cyclopentadecane	C ₁₅ H ₃₀	9-Tricosene, (Z)-	C ₂₃ H ₄₆	Cycloeicosane	C ₂₀ H ₄₀

Cyclotetradecane	C ₁₄ H ₂₈	Cyclododecane	C ₁₂ H ₂₄	Cyclohexadecane	C ₁₆ H ₃₂
Docosane	C ₂₂ H ₄₆	Cyclohexadecane	C ₁₆ H ₃₂	Cyclopentadecane	C ₁₅ H ₃₀
Heptadecane	C ₁₇ H ₃₆	Cyclooctacosane	C ₂₈ H ₅₆	Cyclopropane, 1-methyl-2-octyl-	C ₁₂ H ₂₄
Hexadecane	C ₁₆ H ₃₄	Cyclopentadecane	C ₁₅ H ₃₀	Cyclopropane, 1-methyl-2-pentyl-	C ₉ H ₁₈
Octacosane	C ₂₈ H ₅₈	Cyclotetradecane	C ₁₄ H ₂₈	Cyclopropane, 1-pentyl-2-propyl-	C ₁₁ H ₂₂
Octadecane	C ₁₈ H ₃₈	Docosane	C ₂₂ H ₄₆	Cyclotetracosane	C ₂₄ H ₄₈
Tetracosane	C ₂₄ H ₅₀	Heneicosane	C ₂₁ H ₄₄	Cyclotetradecane	C ₁₄ H ₂₈
Triacontane	C ₃₀ H ₆₂	Henicos-1-ene	C ₂₁ H ₄₂	Dotriacontane, 2-methyl-	C ₃₃ H ₆₈
Z-5-Nonadecene	C ₁₉ H ₃₈	Hentriacontane	C ₃₁ H ₆₄	Eicosane	C ₂₀ H ₄₂
		Heptacosane	C ₂₇ H ₅₆	Heneicosane	C ₂₁ H ₄₄
		Heptadecane	C ₁₇ H ₃₆	Hentriacontane	C ₃₁ H ₆₄
		Hexatriacontane	C ₃₆ H ₇₄	Heptadecane	C ₁₇ H ₃₆
		Nonacos-1-ene	C ₂₉ H ₅₈	Heptadecane, 2,6,10,15-tetramethyl	C ₂₁ H ₄₄
		Octacosane	C ₂₈ H ₅₈	Hexatriacontane	C ₃₆ H ₇₄
		Octadecane	C ₁₈ H ₃₈	Octacosane	C ₂₈ H ₅₈
		Pentatriacontane	C ₃₅ H ₇₂	Tridecane	C ₁₃ H ₂₈
		Tetracosane	C ₂₄ H ₅₀	Z-5-Nonadecene	C ₁₉ H ₃₈
		Triacontane	C ₃₀ H ₆₂	Z-8-Hexadecene	C ₁₆ H ₃₂
		Tricosane	C ₂₃ H ₄₈		
		Z-5-Nonadecene	C ₁₉ H ₃₈		
		Z-8-Hexadecene	C ₁₆ H ₃₂		

4.0 Conclusions

The pyrolysis of the waste HDPE, LDPE, and PP carried out one after the other yielded oil and wax residues in addition to gases given off at a maximum temperature of 510 °C. The yields were all above 50 % in terms of the liquid oil. PP with 58.6 % has the highest oil yields slightly above LDPE with 54.3 % and HDPE with 51.3 % the least obtained value. The S.G was slightly lower than the minimum limit of DPR approved range of 0.820. However, the kinematic viscosity at 40 °C was within the approved range. The flashpoints were found to be below the approved limit. GC-MS analysis showed the oils to contain aliphatic and cyclic hydrocarbons. All three pyrolysis oils produced are suitable for types of diesel blending with conventional diesel. Based on this research project, liquid oil obtained from waste plastics pyrolysis can serve as an alternative new energy resource as well as a mitigating solution to the challenge of plastics waste management. Wastes plastic pyrolysis method is also cost-effective when compared to fossil fuel of non-renewable energy and likewise will contribute to the alleviation of the greenhouse effect.

Further study could be performed to establish the overall efficiency taking into account the effective waste disposal, energy generation, and economic value of all the pyrolysis products including the gaseous, liquid, and solid fractions. However, at the level of this present research, energy efficiency calculation/estimation is not within the scope. The eventual objective of the research to demonstrate an alternative way of addressing plastic waste disposal by converting it to fuel was achieved.

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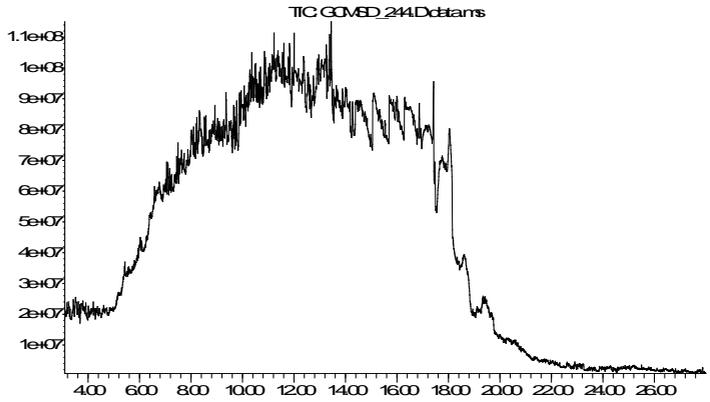
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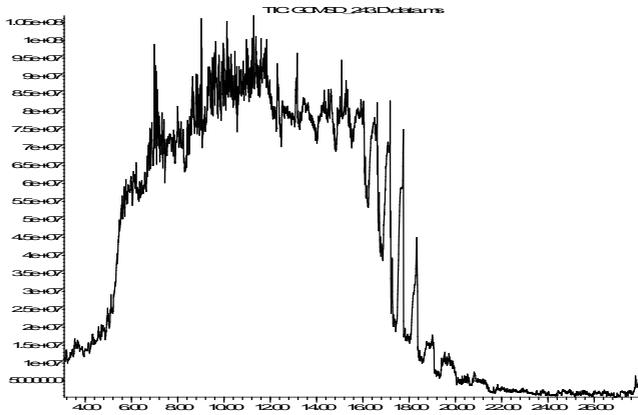
Abundance



Time-->

Figure 2.0: GC-MS Chromatogram for HDPE pyrolysis oil

Abundance



Time-->

Figure 3.0: GC-MS Chromatogram for LDPE pyrolysis oil

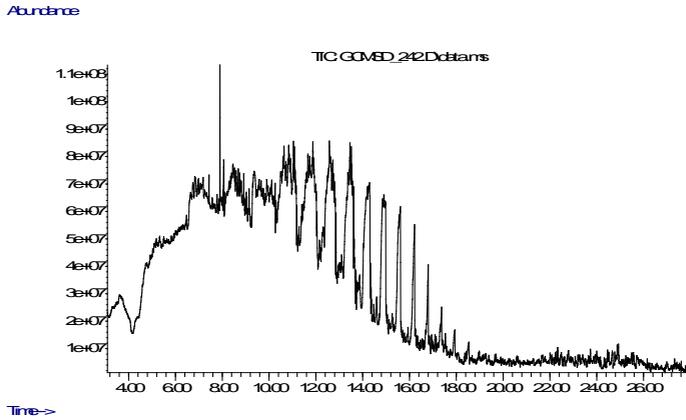


Figure 4.0: GC-MS Chromatogram for PP pyrolysis oil