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**Paper Code: A42**

**Simple Pyrolysis of Waste Plastics made from Polyethylene, Polypropylene and Polystyrene to produce liquid Fuel**

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**Abstract:** *Plastics waste becomes substantial problem for environment, especially in large cities where most inhabitants use plastics extensively. It can be thermally treated by pyrolysis process to produce useful liquid fuel in considerable amount. However, researches must be carried out in order to find the best process variables, such as temperatures, duration, initial mass, etc., and also to obtain the best fuel characteristics, such as kinematic viscosity, specific gravity, flash point, pour point, etc., which are appropriate to internal combustion engines.*

*In order to resemble the real plastics waste which is randomly mixed in waste disposal areas, the research is conducted by varying the proportion of High Density Polyethylene (HDPE), Polypropylene (PP) and Polystyrene (PS), which are mostly used plastics types, as raw materials for pyrolysis. The pyrolysis products are solid, liquid and gas. However, the research focuses only on liquid (oil) products. Those raw materials are pyrolyzed in a cylindrical tank with temperatures between 550 °C and 700 °C. Higher fraction of PS will increase production rates, oil fraction, kinematic viscosity and specific gravity. The maximum oil fraction is 80.72%, while specific gravity is 0.9208. PE as raw material will yield the highest kinematic viscosity and Reid Vapour Pressure, which are 1.422 mm<sup>2</sup>/s and 27.89 kPa, respectively. In terms of Flash and Pour points, oils obtained from PP, PS and their mixtures have the similar values, i.e below 10 °C and below -33 °C. On the contrary, oil from PE has higher pour point (18 °C), yet similar flash point. GC-MS (Gas Chromatography – Mass Spectroscopy) test results show that all oil are roughly 73% - 95% dominated by atomic range of between C<sub>6</sub> and C<sub>12</sub>, therefore they are more appropriate as gasoline fuel.*

**Keywords:** Liquid Fuel, Oil Characteristics, Plastics Mixture, Pyrolysis, Waste Plastics.

## 1. INTRODUCTION

Since decades plastic materials are used intensively for almost all products required by human beings. However, despite of its obvious superiorities as material, such as durability, corrosion resistance, lightweight, formability, low cost, etc., people begin to worry about its low degradation rate after its service life. High production and consumption of plastics leads to a continuous increase of disposed plastic wastes and consequently shortage of landfill areas, since plastics require long time to decay.

Statistics for Western Europe estimated an annual consumption of plastics product at almost 100 kg per person for a total of over 39.1 million tons, and unfortunately around 61% of the generated plastic wastes were simply disposed to landfills [1]. A report from Japan mentioned that in 2006 the domestic plastic wastes had reached a total of 10.06 million tons. Around 72% were reutilized as materials, fuels, electricity or heat [2].

Indonesian Ministry of Environment has released data showing that in 1995 urban inhabitants produced on the average 0.8 kg mixed waste per capita daily. In 2000 the figure increased up to 1 kg/day/person. It is estimated that in 2020 those inhabitants will throw away 2.1 kg/day/person. Assuming that 15% of those mixed refuses are plastics, the 220 million citizens of the country in 1995 disposed approximately 26,500 ton of plastic wastes per day [3]. The figure will look certainly much more terrible in 2012 when both the total inhabitants and the plastics consumption per capita increased considerably.

Most plastic wastes consist of Polyethylene (PE), Polypropylene (PP), Polystyrene (PS), Polyethylene Terephthalate (PET), Polyvinyl Chloride (PVC), Polyurethanes (PU) and Polyamides (PA), which altogether account for more than 90% of total plastics wastes. There are several ways for handling them, e.g. landfilling, melting and regranulation, feedstock recycling, and incineration. Due to some reasons feedstock recycling, such as pyrolysis, appears to be very promising because the obtained valuable chemicals can be used either for raw materials in chemical industry or for fuels [4]. It allows the treatment of mixed, contaminated, or hard to recycle plastic wastes. The production of gasoline, kerosene and diesel oil from waste plastics' pyrolysis is an emerging technological solution to the huge amount of plastics which can not be recovered economically. Mainly PE, PP and PS are used as the feedstock since they have no heteroatom content and theoretically liquid products are free

of sulfur [5]. Pyrolytic recycling of plastic wastes has already been achieved successfully on commercial scale in several plants [2].

## 2. PYROLYSIS OF PLASTIC WASTES

Basically pyrolysis is a decomposition process of plastics by means of heating in an inert atmosphere to yield a variety of products (mostly hydrocarbons) in solid, liquid and gaseous phase. The most significant variables is temperature, because it influences both the polymer conversion and the product distribution, i.e. oils, gases, waxes, and solid residue. As temperature rises, the gas fraction will increase and conversely the liquid fraction will decrease [6,7]. Therefore low and medium temperature pyrolysis, between 400 °C and 500 °C, is used if high fraction of oil is desired.

A variety of reactors have been developed for plastics pyrolysis, such as stirred tanks, shaft furnaces, rotary kilns, fixed beds, fluidized beds, circulating bed reactors and screw extruders [1,4]. Certain pyrolysis may require different type of reactor, because it determines mainly the amount of heat transfer, mixing, residence times and the escape of primary products [5].

Researches must always be carried out in order to find the best process variables, such as temperatures, duration, initial mass, etc., and also to obtain the best fuel characteristics, such as kinematic viscosity, specific gravity, flash point, pour point, etc., which are appropriate to internal combustion engines. Sometimes, if required, plastic wastes are pyrolyzed together with coal [4], lubrication oil wastes [5] or even biomass [6].

## 3. RESEARCH METHODOLOGY

In order to resemble the real plastics waste which is randomly mixed in waste disposal areas, the research is conducted by varying the proportion of High Density Polyethylene (HDPE), Polypropylene (PP) and Polystyrene (PS) as raw materials (feedstock) for pyrolysis [8]. There are 5 samples, i.e. 100% PP, 100% PS, 100% HDPE, 75% PP + 25% PS, and 25% PP + 75% PS. The pyrolysis are carried out three times for each sample with almost similar initial mass, as shown in Table 1, while the following Fig. 1 shows the raw materials used in the research.

Table 1 Initial mass of plastic wastes (in gram)

	Single			Mixed	
	100% PP	100% PS	100% HDPE	75% PP + 25% PS	25% PP + 75% PS
1	255.4	91.2	155.7	161	122.4
2	255.2	97.1	152.4	160.5	120.1
3	253	96.8	152.7	160.5	122.3



Fig. 1: The raw materials: PP (left), PS (middle) and HDPE (right)

Fig. 2 displays the schematic diagram of the simple pyrolyzer unit. Raw materials are placed in a tightly sealed cylindrical feedstock container (D 80 mm, H 480 mm) and pyrolyzed in a cylindrical reactor (stainless steel, D 300 mm, H 500 mm) with temperatures between 550 °C and 700 °C and initial pressure of 1 atm. The high temperature is brought about by a 1200 W heater, which consists of electric resistance wires and controlled with the help of thermocouples. The U stainless tube will direct the pyrolytic gases into Container 1 where some parts condense naturally. The rest which are still in gaseous phase will flow further through a Condenser (D 25 mm, L 48 mm) where some parts condense in Container 2. The remaining gas is then flared to avoid emission of combustibles into the atmosphere. The flow rate of cooling water supplied to the condenser is measured by a flowmeter.

Initially the cylindrical reactor is preheated until roughly 550 °C before the feedstock tube is placed inside. The heating continues until a temperature of around 700 °C. The experiment is terminated if there is no further oil condensation in both containers. Obviously the pyrolysis products are solids (left inside the feedstock tank), liquid (in the container 1 and 2) and flared gas. However, the research focuses only on liquid oil products due to their considerable potential as fuels. The oils are then sent to laboratories for analysis in terms of physical parameters, such as Specific Gravity, Kinematic Viscosity, Flash Point, Pour Point, and Reid Vapour Pressure. Furthermore the oils are analyzed in order to determine their chemical compositions using GC-MS (Gas Chromatography – Mass Spectroscopy).

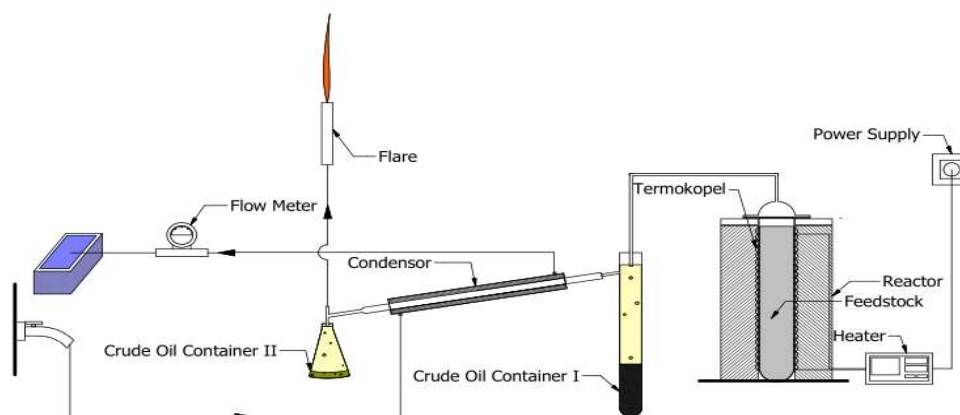


Fig. 2: Schematic diagram of the pyrolyzer

## 4. RESULTS AND DISCUSSIONS

### 4.1 Production rates

The produced oil volume is measured every 10 minutes and the results are shown in Fig. 3. However, the oil yield is taken from the Container 1 only, because the yield in the Container 2 is negligibly small. The poor performance of the Condensor is responsible for it, since the cooling water temperature is practically equal to the ambient temperature (around 27 °C).

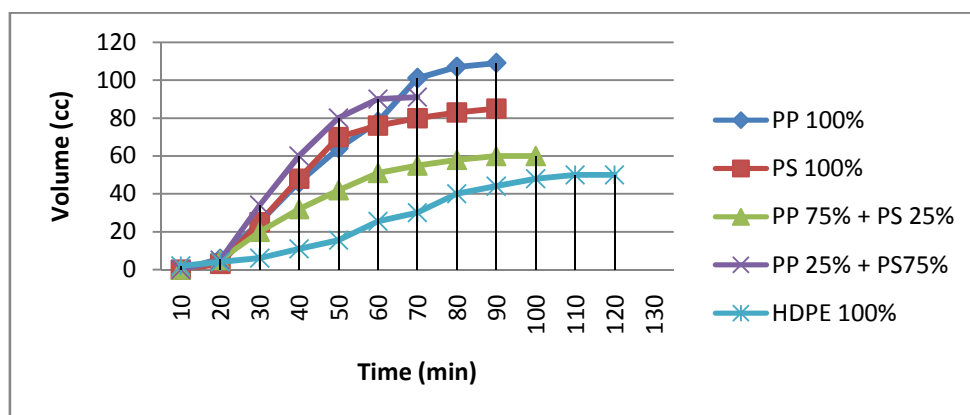


Fig. 3 Oil yield from the Container 1

The figure displays that between 30<sup>th</sup> and 50<sup>th</sup> minutes the curves are mostly linear, therefore very simple calculations can be carried out to determine the instantaneous production rates, as revealed in Table 2. It is obvious that HDPE is the most difficult plastics to pyrolyze. PS is the fastest one to pyrolyze although its initial mass is the lowest (see Table 1). PP behaves between those two other materials. Higher decomposition is observed in PP than in HDPE. It seems that less crystalline or more branched polymers are less stable in thermal degradation [9]. Furthermore, it is observed in the mixture of PS and PP, that higher PS content will yield oil faster than those with lower PS content. If the rates of PP 25% + PS 75% are surprisingly even higher than those of PS 100%, the reason is probably the higher initial mass of the former. The results are in accordance with Lee [10], who observes that pyrolysis of PS shows higher liquid yield than that of polyolefinic polymer, such as PE and PP, because of the structure of stable benzene rings (in PS) instead of straight hydrocarbon structure (in PE and PP). PS is less cracked to gas product of 5 carbon numbers or less. In all cases, the production rates tend to diminish with the elapsed time.

Table 2 Production rates (cm<sup>3</sup>/min)

Time (min)	PP 100%	PS 100%	PP 75% + PS 25%	PP 25% + PS 75%	HDPE 100%
t = 30	2.1	2.3	1.2	2.6	0.5
t = 40	1.95	2.25	1.1	2.3	0.475
t = 50	1.8	2.2	1	2	0.45

#### 4.2 Product composition

After pyrolysis processes at 700 °C of all five samples are fully completed, product compositions in terms of phase (solid, liquid and gas) are analyzed. The results are shown in Table 3.

Table 3 Product composition of the pyrolysis at 700°C (in gram)

No	Feedstocks	Initial mass (g)	Liquid		Gas		Solid	
			Mass	%	Mass	%	Mass	%
1	PP	255.4	97.3	38.1	151.9	59.47	6.2	2.428
2	PS	97	78.3	80.72	15.8	16.28	2.9	2.98
3	PP75%+PS25%	161	58.4	36.27	100.9	62.67	1.7	1.056
4	PP25%+PS75%	120.5	83.7	69.46	32.9	27.3	3.9	3.237
5	HDPE	155.7	44.6	28.64	107.1	68.78	4	2.569

In all cases, the solid fractions are negligibly small. The gas fractions are mostly the highest ones, possibly because of too high pyrolyzing temperature and poor condenser performance, except for the cases where PS is dominant. The maximum liquid yield amounts to 80.72%. Pyrolysis of PS shows higher liquid yield than that of polyolefinic polymer, such as PE and PP, since PS has stable benzene rings, while PE and PS have straight hydrocarbon structure. PS is less cracked to gas product of 5 carbon numbers or less [10]. Higher liquid fraction is observed in PP than in HDPE. It seems that less crystalline or more branched polymers are less stable in thermal degradation [9].

#### 4.3 Oil properties based on its feedstock

The obtained oils are then analyzed in order to identify their physical characteristics, i.e. Specific Gravity, Kinematic Viscosity, Flash Point, Pour Point, and Reid Vapor Pressure. It is not surprising if the waste types determine the physical properties, as shown in Table 4. In all those figures and tables, properties of gasoline, kerosene and diesel fuel are also included just for comparison purposes.

Table 4 Oil properties

Properties	PP	PS	PP25+PS75	PP75+PS25	HDPE	Diesel	Kerosene	Gasoline
Specific Gravity (-)	0.8843	0.9208	0.9072	0.8992	0.7768	0.8445	0.7575	0.75
Kinematic Viscosity (mm <sup>2</sup> /s)	0.95	0.988	0.952	0.867	1.422	4.012	-	0.88
Reid Vapour Pressure (kPa)	25.85	12.24	11.56	16.33	27.89	4.76	-	53.78
Pour Point (°C)	Still liquid at - 33 °C				18	6	-	-
Flash Point (°C)	Already flashed at 10 °C					66.5	25.55 – 57.1	- 56.75

Oil from PS has the highest specific gravity (0.9208), followed by PP-oil and HDPE-oil. Those from their mixture demonstrate their proportionality. HDPE as raw material will yield the highest kinematic viscosity, Reid Vapour Pressure and pour point, which are 1.422 mm<sup>2</sup>/s, 27.89 kPa, and 18 °C respectively. On the other hand, its specific gravity is the lowest. In terms of Flash and Pour points, oils obtained from PP, PS and their mixtures have the similar values, i.e below 10 °C and below -33 °C. However, thorough tests with better equipment are required to determine the exact values. Oil from HDPE has higher pour point (18 °C), yet similar flash point.

#### 4.4 Oil analysis with GC-MS

The conducted analysis shows that the pyrolytic oils contain a lot of chemical substances, which is indicated clearly by many peaks in the GC spectra. Subsequently, based on the available data of retention times, all composing substances can be recognized. Afterward, each substance is examined in a MS instrument in order to determine the relative molecular mass of each substance. Figures 4 – 8 display the chromatograms of all oils. The following tables 5 – 9 show the GC data of the corresponding oils. Finally, tables 10 – 14 reveal the dominant substances of each oil.

There are 52 peaks in the PP-oil (Fig. 4), however the five highest percentages are 14.08% of C<sub>8</sub>H<sub>8</sub>, followed by 11.69% of C<sub>7</sub>H<sub>8</sub>, then 8.12% of C<sub>8</sub>H<sub>10</sub>, next 5.95% of C<sub>9</sub>H<sub>10</sub> and finally 4.03% of C<sub>8</sub>H<sub>10</sub> (Tables 5 and 10).

31 peaks are recorded for the PS-oil (Fig. 5), where the five highest percentages are 38.57% of C<sub>8</sub>H<sub>8</sub>, followed by 10.33% of C<sub>7</sub>H<sub>8</sub>, then 5.34% of C<sub>10</sub>H<sub>8</sub>, next 4.95% of C<sub>20</sub>H<sub>34</sub> and finally 4.12% of C<sub>15</sub>H<sub>24</sub> (Tables 6 and 11).

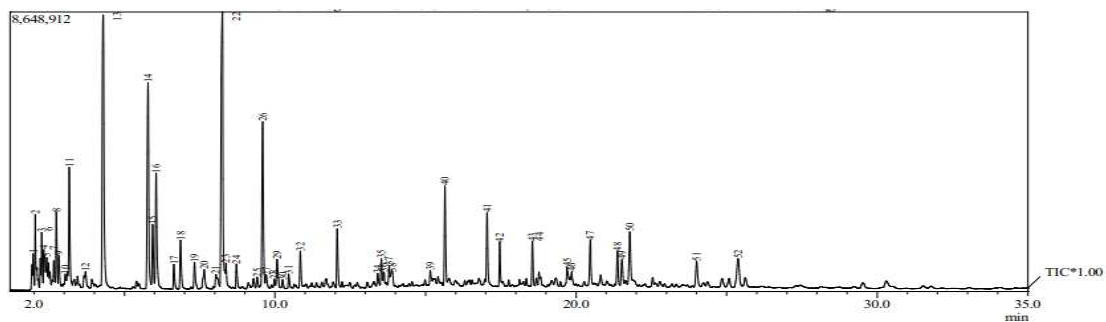


Fig. 4 Chromatogram of the PP-oil

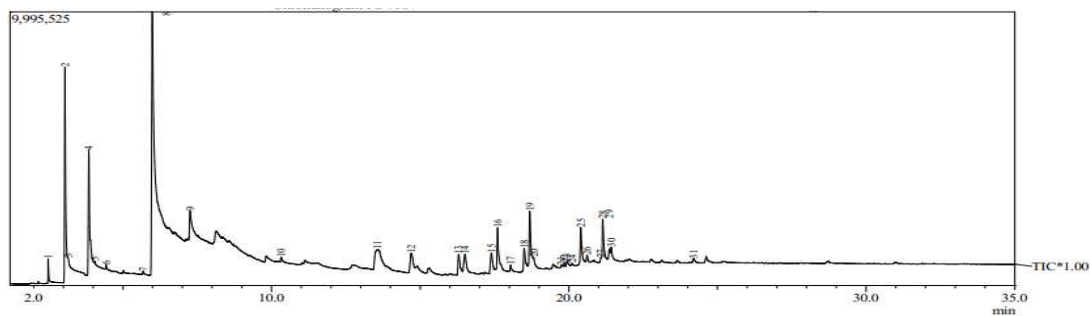


Fig. 5 Chromatogram of the PS-oil

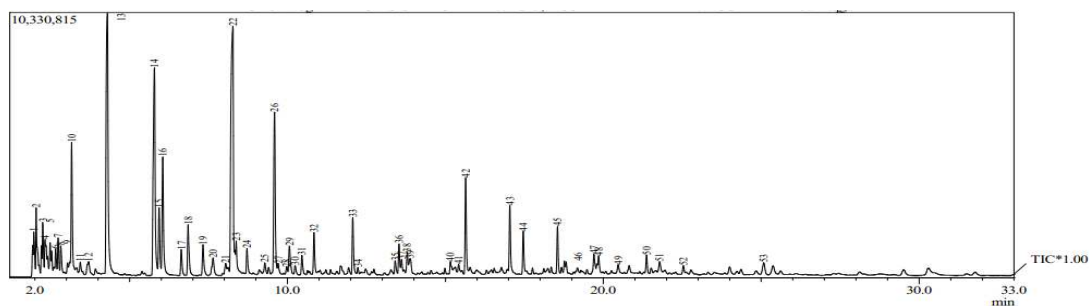


Fig. 6 Chromatogram of the oil from PP 75% + PS 25%

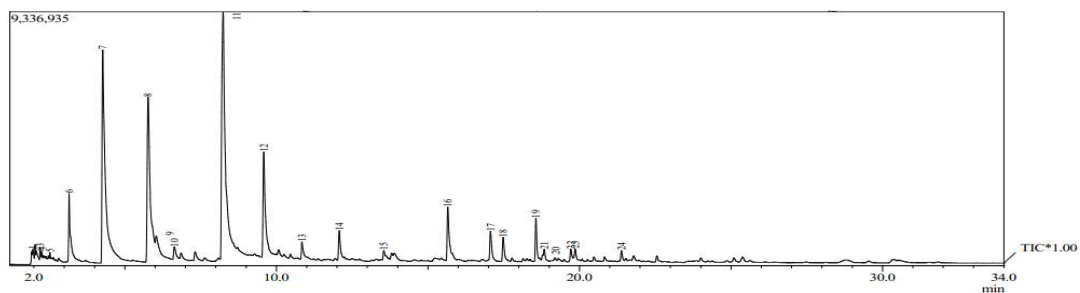


Fig. 7 Chromatogram of the oil from PP 25% + PS 75%

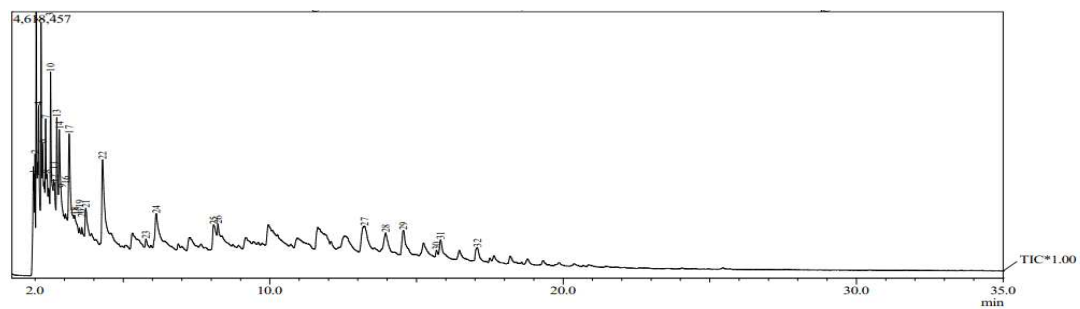


Fig. 8 Chromatogram of the HDPE-oil

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Table 5 GC data of the PP-oil

Peak#	R.Time	I.Time	F.Time	Area	Area%	Peak Report TIC	
						Height	Name
13	4.295	4.200	4.458	31673571	11.69	8373209	
14	5.782	5.692	5.892	22019755	8.12	6361886	
16	6.054	6.000	6.192	10935716	4.03	3538447	
22	8.255	8.167	8.342	38156829	14.08	8515436	
26	9.590	9.475	9.675	16114989	5.95	5137840	

Table 6 GC data of the PS-oil

Peak#	R.Time	I.Time	F.Time	Area	Area%	Peak Report TIC	
						Height	Name
2	3.050	3.017	3.142	19022822	10.33	7800260	
8	5.993	5.933	6.408	71052196	38.57	9372215	
11	13.558	13.408	13.833	9834581	5.34	699017	
16	17.602	17.500	17.842	7580637	4.12	1625868	
19	18.686	18.608	18.800	9117123	4.95	2145104	

Table 7 GC data of the oil from PP 75% + PS 25%

Peak#	R.Time	I.Time	F.Time	Area	Area%	Peak Report TIC	
						Height	Name
10	3.165	3.117	3.275	13360327	3.92	5021931	
13	4.297	4.192	4.517	44996505	13.22	10158923	
14	5.780	5.675	5.883	31988799	9.40	8076992	
22	8.268	8.158	8.317	51645160	15.17	9595401	
26	9.588	9.492	9.667	20894651	6.14	6277679	

Table 8 GC data of the oil from PP 25% + PS 75%

Peak#	R.Time	I.Time	F.Time	Area	Area%	Peak Report TIC	
						Height	Name
7	4.271	4.208	4.733	53806691	21.56	7801758	
8	5.763	5.683	5.992	42149640	16.89	6045486	
11	8.241	8.150	8.667	73648840	29.50	9035233	
12	9.583	9.517	9.858	20205041	8.09	3854021	
16	15.654	15.600	15.792	7721553	3.09	1899447	

Table 9 GC data of the HDPE-oil

Peak#	R.Time	I.Time	F.Time	Area	Area%	Peak Report TIC	
						Height	Name
4	2.113	2.067	2.175	11168637	7.19	2859990	
7	2.356	2.283	2.383	10010118	6.44	2548514	
10	2.526	2.500	2.608	11409324	7.34	3300543	
14	2.824	2.783	2.958	12835183	8.26	2197914	
17	3.160	3.092	3.267	10981744	7.06	2011099	

Table 10 Dominant substances in the PP-oil

No	Peak	% Area	Substance	Formula
1	13	11.69	Toluene/Methylenzene	C <sub>7</sub> H <sub>8</sub>
2	14	8.12	Ethylbenzene	C <sub>8</sub> H <sub>10</sub>
3	16	4.03	1,4 Dimethylbenzene	C <sub>8</sub> H <sub>10</sub>
4	22	14.08	Styren/Benzene ethenyl	C <sub>8</sub> H <sub>8</sub>
5	26	5.95	Isoallyenzene	C <sub>9</sub> H <sub>10</sub>

Table 11 Dominant substances in the PS-oil

No	Peak	% Area	Substance	Formula
1	2	10.33	Toluene/Benzene-Methyl	C <sub>7</sub> H <sub>8</sub>
2	8	38.57	Styren/EthenylBenzene	C <sub>8</sub> H <sub>8</sub>
3	11	5.34	Napthalene/Azulene	C <sub>10</sub> H <sub>8</sub>
4	16	4.12	Cis-Caryophyllene	C <sub>15</sub> H <sub>24</sub>
5	19	4.95	Cyclohexane	C <sub>20</sub> H <sub>34</sub>

Fig. 6 shows 53 peaks in the oil of PP75%+PS25%, while Tables 7 and 12 reveal the five highest percentages, i.e. 15.17%, 13.22%, 9.4%, 6.14% and 3.92% for C<sub>8</sub>H<sub>8</sub>, C<sub>7</sub>H<sub>8</sub>, C<sub>8</sub>H<sub>10</sub>, C<sub>9</sub>H<sub>10</sub> and C<sub>6</sub>H<sub>6</sub>, respectively.



There are 24 peaks in the oil of PP25%+PS75% (Fig. 7), however the five highest percentages are 29.5% of C<sub>8</sub>H<sub>8</sub>, followed by 21.56% of C<sub>7</sub>H<sub>8</sub>, then 16.89% of C<sub>8</sub>H<sub>10</sub>, next 8.09% of C<sub>9</sub>H<sub>10</sub> and finally 3.09% of C<sub>10</sub>H<sub>8</sub> (Tables 8 and 13).

As identified in Fig. 8, HDPE-oil has 32 peaks are identified, but only the five highest percentages are considered, i.e. 8.26% of C<sub>12</sub>H<sub>26</sub>O, followed by 7.34% of C<sub>8</sub>H<sub>16</sub>, then 7.19% of C<sub>7</sub>H<sub>16</sub>, next 7.06% of C<sub>11</sub>H<sub>22</sub> and finally 6.44% of C<sub>8</sub>H<sub>18</sub> (Tables 9 and 14).

Table 12 Dominant substances in the oil of PP 75% + PS 25%

No	Peak	% Area	Substance	Formula
1	10	3.92	Benzene	C <sub>6</sub> H <sub>6</sub>
2	13	13.22	Benzene Methyl	C <sub>7</sub> H <sub>8</sub>
3	14	9.40	EthylBenzene	C <sub>8</sub> H <sub>10</sub>
4	22	15.17	Benzene Ethenyl	C <sub>8</sub> H <sub>8</sub>
5	26	6.14	Alpha MethylStyrene	C <sub>9</sub> H <sub>10</sub>

Table 13 Dominant substances in the oil of PP 25% + PS 75%

No	Peak	% Area	Substance	Formula
1	7	21.56	Benzene methyl	C <sub>7</sub> H <sub>8</sub>
2	8	16.89	Ethylbenzene	C <sub>8</sub> H <sub>10</sub>
3	11	29.50	Styren/BenzeneEthenyl	C <sub>8</sub> H <sub>8</sub>
4	12	8.09	Alpha Methyl Styrene	C <sub>9</sub> H <sub>10</sub>
5	16	3.09	Nepthalene	C <sub>10</sub> H <sub>8</sub>

Table 14 Dominant substances in the HDPE-oil

No	Peak	% Area	Substance	Formula
1	4	7.19	Heptane	C <sub>7</sub> H <sub>16</sub>
2	7	6.44	Octane	C <sub>8</sub> H <sub>18</sub>
3	10	7.34	Octane Caprylene	C <sub>8</sub> H <sub>16</sub>
4	14	8.26	1-Octanol,2-Butyl	C <sub>12</sub> H <sub>26</sub> O
5	17	7.06	Undecene	C <sub>11</sub> H <sub>22</sub>

If most of those hydrocarbon substances are classified into 2 main groups, where the first contains of C-6 until C-12, while the second between C-13 and C-23, the GC-MS test results reveal an interesting phenomenon, as seen in Table 15.

Table 15 Two main groups of carbon fractions in the oils

No	Feedstock	Group of carbon fractions		Most dominant carbon fraction
		C <sub>6</sub> - C <sub>12</sub>	C <sub>13</sub> - C <sub>23</sub>	
1	PP	85.65	10.55	C <sub>8</sub> = 31.83%
2	PS	73.83	21.8	C <sub>8</sub> = 47.55%
3	PP 75% + PS 25%	93.47	4.42	C <sub>8</sub> = 34.92%
4	PP 25% + PS 75%	95.63	2.43	C <sub>8</sub> = 49.01%
5	HDPE	84.19	8.18	C <sub>7</sub> = 25.02%

Obviously, all oils are roughly 73% - 95% dominated by the atomic range of between C<sub>6</sub> and C<sub>12</sub>. The most dominant carbon fraction is C<sub>8</sub> as well, except that in the HDPE oil. Therefore they are more appropriate as gasoline fuel.

## 5. CONCLUSIONS

1. Pyrolysis is technically feasible to convert plastic wastes to useful liquid fuel.
2. PS is the best feedstock in terms of liquid yield rate and percentage. On the other hand HDPE is the worst one, while PP is in between.
3. In general, mixture of PP and PS wastes can improve the pyrolysis characteristics of pure PP, but understandably worsen those of pure PS.
4. Specific Gravities of oils from PP, PS and their mixtures are close to (slightly above) diesel fuel, while oil from HDPE has the lowest value and closer to gasoline.
5. Kinematic Viscosities of oils from PP, PS and their mixtures are almost similar to gasoline, while HDPE-oil has the highest value, however it is still too low compared to diesel fuel.
6. Those oils are more appropriate if used as gasoline rather than diesel fuel.



7. Higher yield and better quality of the liquid fuel can be achieved by improving and optimizing the process, e.g. appropriate temperature, addition of catalytic reforming, lower cooling water temperature, etc.

## **6. ACKNOWLEDGEMENTS**

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