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Non-catalytic Hydrothermal Conversion of Individual and Mixed Plastic Waste to Hydrocarbon Fuels and Chemicals

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Abstract: The global plastic production rate exceeds 430 million tons annually, and about five billion tons of these plastic wastes have been accumulated in landfills and the natural environment. The polypropylene, polystyrene, and polyethylene terephthalate account for about 30.4% of the total waste. Hydrothermal co-processing of mixed plastic feedstocks could offer the advantage of potential synergistic reaction environments for producing liquid products of high fuel quality. This study examines the non-catalytic hydrothermal conversion of polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and their mixtures, which were investigated at 400 °C for 2 h under autogenous pressure. The hydrothermal products of PP and PS are predominantly liquids with 73.65 and 83.58 wt.% yields, respectively, with low gas and hydrochar generated. The PET produced hydrochar (47.28 wt.%) as the dominant product, generating only 28.60 wt% oil. Results from the hydrothermal co-processing of mixed plastic show that synergistic interactions occurred between the degradation products of the plastics, with an oil yield of 66.05 wt.% and hydrochar of 15.71 wt.%. The GC-MS analyses of the liquid products indicated the presence of paraffinic, olefinic, and aromatic hydrocarbons. The PS and PET oils are dominated by aromatic hydrocarbons with 1,3-diphenylpropane (19.20%) and 1,4-Benzenedicarboxylic acid, diethyl ester (30.42%), while PP is rich with paraffinic and olefinic compounds, accounting for 44.42 and 41.48 %, respectively. These results demonstrate effective hydrothermal conversion of mixed post-consumer plastics into high-quality liquid fuels and aromatic chemicals under moderate reaction conditions.

1. Introduction

Plastics have become an integral part of modern lifestyle due to their versatility, durability, cost-effectiveness, convenience, high transparency, superior chemical stability, barrier properties, and mechanical properties. Plastic materials are widely employed in a variety of industries, including food packaging, plastic films, electronic devices, and mechanical equipment (Su et al., 2023; Yang et al., 2021; Gorjizajeh et al, 2017). Global plastic usage has been steadily increasing annually in recent years, driven by ongoing economic expansion and the enormous demand for plastic products, as well as their relatively short lifespan. A lot of plastics are thrown away as waste, which negatively impacts the environment and wastes resources if not treated properly (Yang et al., 2023; Pilapitiya and Ratnayake,

2024). About 1.2 billion tons of plastic waste are predicted to be dumped in landfills or released into the environment by 2050 due to ineffective recycling systems (Geyer et al., 2017; Su et al., 2023). Synthetic plastics, such as polypropylene (PP), polyethylene (PE), polystyrene (PS), polyethylene terephthalate (PET), polyvinyl chloride (PVC), and polyurethane (PU, PUR), are generally derived from the petrochemical industry, and classified as high molecular weight materials and mostly contain branch and crosslinked polymer structures (Khumthai et al., 2022; Elidrissi et al., 2007). These polymers are extensively utilized across various industries to replace natural materials in the manufacturing of consumable or disposable products due to their stability, ease of production, cost-effectiveness, and convenience. Despite these advantages, plastics pose challenges in terms of recycling, leading to the substantial accumulation of waste plastics in landfills and ecosystems, causing significant environmental issues (Amobonye et al., 2021; Osman et al., 2018). Conventional methods, including incineration, mechanical recycling, and pyrolysis, are ineffective in reducing plastic waste. Incineration releases greenhouse gases that are toxic and have low energy recovery (Jin et al., 2021; Onwudili & Williams, 2023; Verma et al., 2018). Mechanical recycling of mixed waste typically yields dark-coloured, lower-value products (Jin et al., 2021; Nduwimana et al., 2026). Pyrolysis can convert mixed plastic waste into oils with significant yields (50 to 90%), but generates significant amounts of polycyclic aromatic hydrocarbons that require further processing (Akubo et al., 2019; Hadi et al., 2017; Muhammad et al., 2015; Park et al., 2019). Gasification converts mixed plastic waste into gases such as CH_4, H_2, CO and CO_2 (Jin et al., 2021; Sokoto et al., 2020) with high energy consumption up to 800 ◦C and loss of polymer structure or carbon chain (Bai et al., 2020; Emembolu et al., 2022). Therefore, the search for suitable treatment procedures to directly transform mixed plastic wastes into clean fuels and valuable chemicals may streamline the negative effects caused by the conventional techniques of mixed plastic waste recycling and enhance the sustainability of the recycling process. Hydrothermal conversion processes have recently been regarded as an efficient method for converting mixed plastic waste into compounds with added value under mild reaction conditions (Joshi & Vaidya, 2024; Ling et al., 2023; Onwudili & Williams, 2023). Hydrothermal conversion is a thermochemical process done using subcritical and supercritical water to produce liquid products from feedstock, where water can be a solvent, reactant, or catalyst in the reaction at moderate temperature (280-380 ºC) and pressure (7-30MPa) for the depolymerization process in a batch enclosed reactor. However, hydrothermal liquefaction (HTL) is highly suitable for processing wet organic wastes and materials, thereby saving substantial energy costs from the intensive drying required by pyrolysis. In addition, HTL uses a hot-pressurised water medium, which has unique properties that promote fast reaction and mass transfer rates (Lachos-Perez et al., 2022; Yüksel Özşen, 2020).

This work investigated the hydrothermal conversion of three individual plastics and their plastic mixture. The reactions were carried out in a batch reactor under a reaction temperature of 400 ºC, 2h reaction time and autogenous pressure. The yields of products and detailed analyses of oil products were used to evaluate the synergistic effects from the co-hydrothermal conversion of the mixed plastics, leading to the formation of hydrocarbon-rich oil products.

2. Methodology

2.1 Sourcing and Preparation of Sample

The plastic samples collected for the hydrothermal conversion experiments are waste polystyrene (PS), polypropylene (PP), and polyethylene terephthalate (PET), which were obtained from Usmanu Danfodiyo University's main campus in Sokoto. The waste plastics were cut into small pieces with the aid of scissors.

2.2 Experiments

Hydrothermal conversion was carried out according to the modified procedure described by Onwudili and Williams (2023). A 250 cm^3 stainless steel batch reactor (GSH-25080701 model) was used for the Hydrothermal conversion of plastic waste. It was controlled with a PID AI-509 reactor controller. The reactor can withstand pressures of 30 MPa and temperatures of up to 500 ºC. A typical reaction was performed using the following procedure: about 10 g of the plastic waste sample and 70 g of deionized water were placed into the reactor. The reactor was subsequently closed and locked with a torque wrench to ensure even pressure distribution, followed by flushes with Nitrogen to purge any residual air/oxygen. The reactor was heated to the target reaction temperature of 400 ºC under mechanical stirring at 400 rpm, with a heating rate of approximately 10 ºC per minute and the reactor was maintained at that temperature for 2 hours. The reactor was cooled to a temperature below 30 ºC.

2.3 Product Separation and Yield Determination

The gaseous product from the reactor was collected in a gas bag, while the products in the reactor were recovered using methylene chloride and deionized water (1:1). The solid products were recovered using Whatman No. 1 filter paper and the filtrate was centrifuged out to separate the oil-water mixture. The methylene chloride was recovered using a rotary evaporator at 40 ºC. The yield of products is calculated according to equation 1-4 (Chien et al., 2025; Jin et al., 2021).

$$\boldsymbol{Oil\ yield}\ (\boldsymbol{wt}\%) = \frac{\boldsymbol{Weight\ of\ Oil}}{\boldsymbol{Weight\ of\ Feedstock}} \times \mathbf{100} \qquad (1)$$

$$\boldsymbol{Hydrochar}\ (\boldsymbol{wt}\%) = \frac{\boldsymbol{Weight\ of\ dried\ Residue}}{\boldsymbol{Weight\ of\ Feedstock}} \times \mathbf{100} \qquad (2)$$

$$\boldsymbol{Gas\ Yield + Mass\ loss}(\boldsymbol{wt}\%) = \left(\mathbf{1} - \boldsymbol{Oil_{Yield}} - \boldsymbol{Hydrochar_{yield}}\right) \times \mathbf{100} \qquad (3)$$

$$\boldsymbol{Conversion}\ (\boldsymbol{wt.}\%) = (\mathbf{1} - \boldsymbol{Solid\ Yield}) \times \mathbf{100} \qquad (4)$$

2.4 Product Analysis

An Agilent 6890N GC system was used for the analysis of samples collected during non-catalytic hydrothermal experiments. The instrument was fitted with a 5973N MS detector, auto-injector and a 30 m long DB - 5MS capillary column with an internal diameter of 250 µm and film thickness of 0.25 µm. The carrier as used was helium (99.999%) at a flow rate of 1 mL/min^{-1}. A split ratio of 100:1 and injection volume of 0.5 µL were selected as part of the GC method. Both injector and detector temperatures were maintained at 300 ºC. A ramp method was developed to separate all the components in the sample. In the ramp method for the oil analysis, the oven temperature was initially set at 50 ºC, and the sample was then injected by the auto injector. The oven temperature was maintained at 50 ºC for 4 min after the sample was injected and ramped from 50 ºC to 300 ºC at a rate of 20 °C min^{-1}. Each sample takes 16 min to be analysed by GC, and the oven temperature was cooled back to 50 ºC before the next run was started.

3. Results and Discussion

3.1 Hydrothermal Conversion Products

The product yields from the hydrothermal conversion of individual plastics, PP, PS, and PET and their mixture (PP:PS:PET), are presented in **Figure 1** as percentages of oil, gas, and hydrochar.

Polystyrene (PS) exhibits the highest oil selectivity at 83.58 wt.%, with minimal gas (6.63 wt. %) and hydrochar (9.79 wt.%). The highest oil yield of PS could be attributed to β-scission of aromatic

backbone depolymerisation under hydrothermal conditions to form mono and poly aromatic liquids. Additionally, the resonance stability of intermediate products could suppress secondary cracking and repolymerization that lead to more liquid products than gases and hydrochar. Seshasayee and Savage (2020) reported an oil yield of 86 wt.% at supercritical conditions (450 °C, 0.5–1 h).

As shown in Figure 1a, polypropylene (PP) has a moderate oil yield of 73.65 wt.%, 14.09 wt.% gas, and 12.26 wt.% hydrochar relative to PS, which could be due to the saturated aliphatic structure of the PP that influences random chain scission and hydrogen-deficient radical pathways that promote secondary cracking and condensation reactions. Zhao et al. (2018) showed a 36 wt.% conversion of the PP at 350 °C extending the reaction time to an hour. The same study by Jin et al. (2021) with modified hydrothermal liquefaction (HTL) procedure showed that temperature, and thus the radical mechanism, is more influential on the degradation of PP, gaining an 89 wt.% oil yield at 450 °C for 45 minutes. Mukundan et al. (2022) conducted HTL of PP alone at the subcritical conditions yielded mainly 58.6%, 50.2% and 47.4% solid residue products, with an oil yield of 15.2, 16.5 and 17.2% at 340 °C, 360 °C, and 380 °C, respectively.

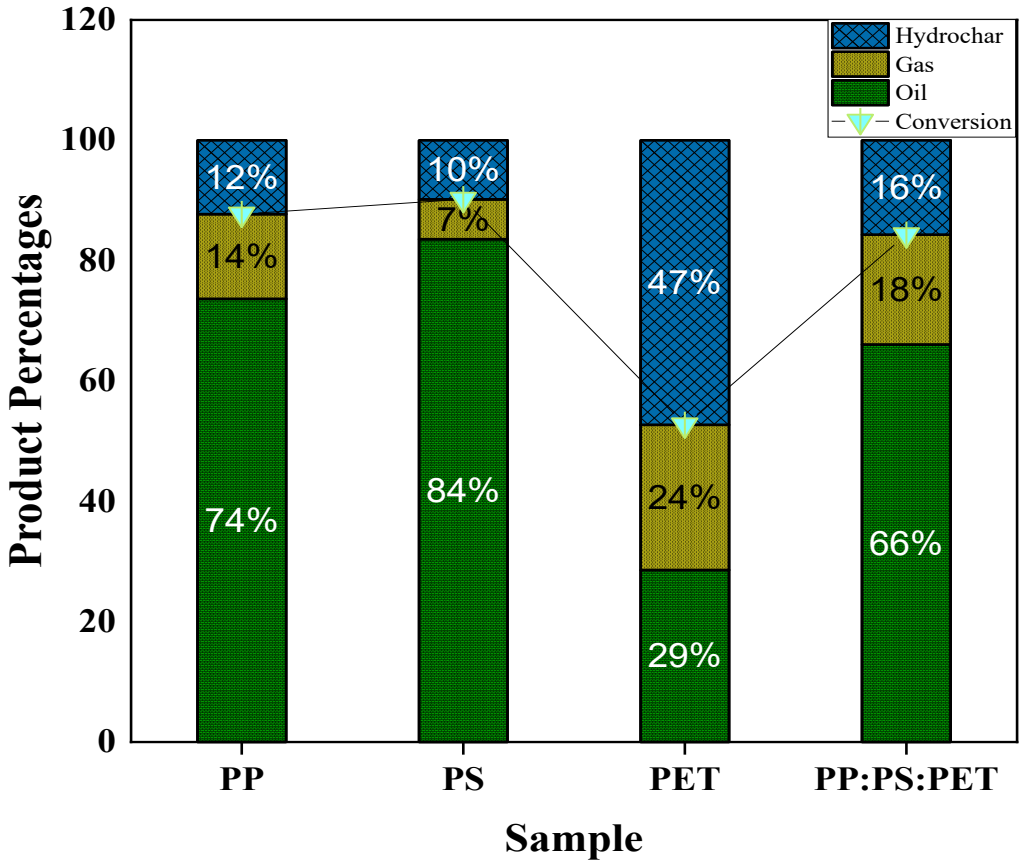

Figure 1. Percentage yield of oil, gas and hydrochar from non-catalytic hydrothermal conversion at 400 °C, 2 h reaction conditions.

In contrast, PET shows (**Figure 1a**) the lowest oil yield (28.60 wt.%) and the highest hydrochar (47.28 wt.%). This is due to its ester backbone and aromatic ring structure, which favour hydrolysis, decarboxylation, and polycondensation reactions that produce oxygen-rich intermediates converting to hydrochar via dehydration. A 10 wt.% oil yield was reported by dos Passos et al. (2020) with a 70% solid yield of pure terephthalic acid (TA) after 30 minutes. PET fully decomposes into its substituent monomers at both subcritical and supercritical temperatures, although the solid TA yield decreases with increasing temperature (Boel et al., 2024). Williams and Slaney (2007) conducted hydrothermal

liquefaction of polyethene terephthalate under hydrogen conditions; the yields were 41 wt.% solid residue and 27 wt.% oil. Ethylene glycol further decomposed into acetaldehyde gas and small alkyls and alkenes through dehydration, which could then form long-chain fatty acids and diols through small-scale oligomerization, while TA formed benzoic acid at higher temperatures due to decarboxylation. Seshasayee and Savage (2020) reported the highest energy recovery (33%) in oil from PET under hydrothermal conditions of 450 °C and a residence time of 1h.

The mixed plastic feedstock shows a significant oil yield of 66.05 wt.%, 18.24 wt.% gas and 15.71 wt.% hydrochar (**Figure 1a**), highlighting a synergistic effect derived from PS aromatic radicals that inhibit char formation from PET. Likewise, PET influences gas formation through secondary cracking and decarboxylation reactions.

3.2 Compositional Analysis of the Hydrothermal Products

The chemical composition of the produced oil from the hydrothermal conversion of PP at 400 °C and 2 h reaction time are presented in **Table 1.**

Table 1. GC-MS profile of PP products from hydrothermal conversion

SN	Name	%Peak Area	Retention Time (Min)
1	2,3-dimethylhept-3-ene	15.47	5.309
2	Hexadecane	15.21	6.814
3	1-Hexacosene	9.66	7.472
4	Octyl-cyclopropane	8.69	5.704
5	1,2-Diethyl-1-methylcyclohexane	7.52	3.964
6	5-methyldocos-5-ene	6.26	8.376
7	1-butyl-2-propylcyclopentane	5.98	6.459
8	Tetradecane	5.76	4.165
9	4,8-Dimethylnona-1,7-diene	4.91	4.416
10	1,3-Diphenylpropane	4.00	7.180
11	17-Pentatriacontene	2.76	6.299
12	Eicosane	1.26	11.317
13	7-methyl-1-undecene	1.24	4.783
14	Octadecene	1.18	9.950
15	Others	10.1	-

The GC–MS profile and chemical distribution obtained from of PP hydrothermal conversion (**Table 1**) reveals the presence of aliphatic hydrocarbons, including branched alkenes (2,3-dimethylhept-3-ene), linear alkanes (hexadecane, tetradecane, eicosane), long-chain olefins (1-hexacosene, 17-pentatriacontene), and cycloalkanes (alkyl-substituted cyclopentane, cyclohexane, and cyclopropane derivatives) with 2,3-dimethylhept-3-ene 15.47%) and Hexadecane (15.21%) as the dominant compounds. The products are formed due random C-C and β-scission reactions as a result of

low enthalpy of C–CH_3 (335 kJ mol^{-1}) bonds, lower than that of the main chain C–C bonds (347–377 kJ mol^{-1}), which caused methyl side groups in PP to readily split from the main chain during degradation, causing higher rates of β-scission and providing a larger alkane-to-alkene ratio throughout the degradation. Likewise, the splitting of methyl side groups also enhances the conversion into alcohols through the interaction with water, which can be dehydrated back to alkenes at higher temperatures, which then allows for a much higher formation of cyclics compounds (Boel et al., 2024). Zhao et al. (2018) analyzed the chemical composition at 400 °C, where 75% of the oil was olefin, with a 5% paraffin and 14% cyclic. Similarly, Chen *et al.* (2019) showed that longer reaction times at 425 °C caused further conversion of alkenes into cyclics and aromatics, olefins decreasing from 93% to 49% with reaction times increasing from 30 minutes to 2 hours. Seshasayee and Savage (2020) reported that from 400 °C and up, cyclics and aromatics were the most prominent components in the oil, with a 40% aromatic proton fraction in ^{1}H NMR, and at 450 °C almost 70% of the lighter oil was composed of multicyclic aromatics like naphthalene and fluorene. The chemical composition of the oil derived from hydrothermal conversion of PS are presented in **Table 2**.

Table 2. GC-MS Profile of PS Products from Hydrothermal Conversion

SN	Name	%Peak Area	Retention Time (Min)
1	1,3-diphenylpropane	19.20	7.209
2	4-Phenyltetracyclonon-4-ene	12.79	7.587
3	Hexadecane	3.67	6.814
4	Benzaldehyde	8.48	10.282
5	1-phenyl-3-methylpropene	3.33	7.764
6	5-methylcycloheptene	4.89	8.657
7	Benzoic acid	2.34	4.720
8	1-phenylnaphthalene	3.67	8.170
9	n-hexadecanoic acid	1.74	8.428
10	tetrahydromethanophthalene	4.05	9.149
11	1,3-diphenylbenzene	4.09	9.452
12	2,4-dimethylbenzene	5.60	10.654
13	m,m-terphenyl derivatives	9.86	11.111
14	1,3,5-triphenylcyclohexane	0.76	12.771
15	Others	15.53	-

Musivand et al. (2023) reported that the decomposition of PS in an HTL environment follows a free radical chain mechanism that involves three processes, including chain initiation, propagation and termination. The olefin lateral chains containing the double bond were more unstable and tended to form radicals that reacted with each other and produced aromatics with more rings, and the polycyclic aromatics are formed by a combination of radicals, cyclisation reactions or intermolecular

rearrangement. Therefore, hydrothermal process appears to be highly effective for the production of renewable aromatics, which accounted for more than 70% of the percentage area of the GC-MS list of compounds in the form of alkane, alkene substitutes and aromatic species with 1, 2 or 3 rings. The chemical composition of the oil derived from hydrothermal conversion of PET is presented in **Table 3.**

Table 3. GC-MS Profile of PET Products from Hydrothermal Conversion

SN	Name	%Peak Area	Retention Time (Min)
1	Benzoic Acid	28.60	4.851
2	Benzoic acid methyl esters	8.87	4.113
3	1,3-Benzenedicarboxylic acid, diethyl ester	30.42	6.150
4	Dimethyl terephthalate	14.82	6.408
5	Hexadecanoic acid	2.29	8.393
6	5-octadecene	2.58	5.796
7	Hexadecane	5.42	6.825
8	Others	9.29	-

The product resulting from PET hydrothermal conversion consists primarily of aromatic oxygenated compounds (82.71%). Paraffins (7.71%) and olefins (2.58%) constitute minor fractions (**Table 3**). The main compounds identified include benzoic acid, benzoic acid methyl ester, 1,4-benzenedicarboxylic acid diethyl ester, and dimethyl terephthalate. In hydrothermal conversion, a process where water at high temperature and pressure is used to break down materials, PET undergoes hydrolytic cleavage, meaning water breaks its ester linkages, which regenerates its monomers under neutral subcritical conditions as well as the presence of oxygen as heteroatoms contributes greatly to depolymerization reactions (dos Passos et al., 2020). The presence of terephthalate esters, such as dimethyl and diethyl terephthalate, likely results from transesterification, a reaction where esters exchange their alkoxy group with an alcohol present in the medium. This typically occurs in non-catalytic hydrothermal conversion, where trace amounts of alcohols or moisture-derived dehydration products act as reactants (Mihajlovic *et al.,* 2025). Teoh et al. (2025) performed co-liquefaction of PET with palm empty fruit bunch, and the GC-MS analysis revealed that the relative abundance of phenolic compounds increased from 28 % to 38 % with an increase in PET content from 20 wt.% to 60 wt.% in the feed mixture the biocrude oil having significant higher ester content, up to 82.7 % for pure PET and 25.8 % for a feed containing 20 wt.% PET. The chemical composition of the oil derived from hydrothermal conversion of mixed PP, PS and PET are presented in **Table 4.**

The results of the mixed PP:PS:PET hydrothermal conversion reveal a synergistic relationship between compounds derived from PP, PS, and PET (Table 4), with PS/PET-derived benzoic acid having the highest relative peak area (53.90%). Other aromatics that are similarly derived from PS,

such as diphenyl cyclopropane, terphenyl derivatives, and the presence of significant amounts of paraffinic and olefinic hydrocarbons (11.61 and 7.59 %) is due to the C-C and β-scission of PP that produces more isoparaffins and olefins due to its methyl branches, which subsequently convert into cyclics and aromatics (Boel et al., 2024). Seshasayee and Savage (2021) performed HTL on an equal mixture of PET, PS, PP, and PC, showing significant synergy at 300 °C, with the polyolefins primarily decomposed into paraffins, olefins, cyclics, and aromatics, and highlighted that the ratio of paraffin to olefin hydrocarbons from the mixed plastics depended on the presence of side chains in the polymer, with PP and (HI)PS having a significantly larger olefin to-paraffin ratio than the rest of the polymers.

Table 4. GC-MS Profile of Mixed Plastics (PP:PS:PET) Products from Hydrothermal Conversion.

SN	Name	%Peak Area	Retention Time (Min)
1	Benzoic acid	53.90	5.057
2	1,1-diphenyl-2-methylpropane	6.47	7.329
3	n-hexadecanoic acid	4.23	8.416
4	hexadecane	7.29	6.820
5	2-cyclopentenyl-1-propene	3.86	10.047
6	1,2-dimethyl-3-butenylbenzene	2.27	10.213
7	1,2-diphenyl cyclopropane	3.24	7.552
8	1,3-dimethylpropene	3.73	7.844
9	Tetradecane	3.41	4.165
10	10-cyclohexylnonadecane	0.52	13.652
11	Cyclotetradecane	0.39	12.759
12	m,m-terphenyl derivatives	2.45	11.111
13	Others	8.24	-

Chand et al. (2022) attempted an HTL of microplastics coming from sewage sludge produced by a wastewater treatment plant in a continuous reactor, and the products showed a 97 wt.% loss of microplastics with 25 wt.% oil recovery, with most of the decomposed microplastics being found in the solid residue. Ciuffi et al. (2021) performed HTL at 340 °C for 5 hours of collected waste directly from a recycling plant, containing cellulose from paper, small amounts of metal, and plastics, PET, PS, PVC, PE, and PP and yielded a 7.7 wt.% oil yield and an 11 wt.% gas yield, with the remaining product being solid. Analysis of the oil showed significant PET and some PS degradation, but no decomposition products of the polyolefins PE and PP.

3.3 Hydrocarbon Distribution

The hydrocarbon distribution of the oil obtained from individual and mixed plastic hydrothermal conversion are illustrated in **Figure 2**. The oil from PP is dominated by paraffinic (44.42 wt.%) and olefinic (41.48 wt.%) indicating that PP undergo free radical mechanism from random β-scission and hydrogen abstraction or recombination (**Figure 2**). The significant increase in the paraffins in the oil could be due to in-situ hydrogenation facilitated by hydrogen from the solvent, while the low aromatic (4.0 %) content revealed minimal cyclisation and aromatisation at the reaction conditions. Polystyrene oil is primarily dominated by the aromatic (74.17%) with low paraffinic and olefinic

hydrocarbons due to the lack of a long aliphatic carbon chain. The higher aromatic in the oil is favoured by the polystyrene unzipping reaction to produce monomers, dimers, and polycyclic aromatic compounds.

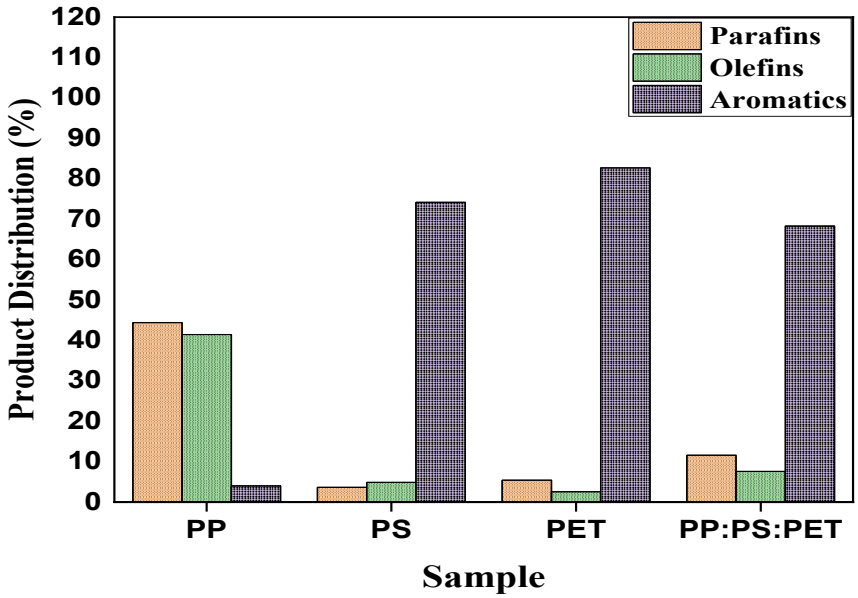

Figure 2. Hydrocarbon product distribution from PP, PS, and Mixed plastic hydrothermal conversion at 400 °C, 2 h.

The hydrothermal treatment of PET involves hydrolysis to terephthalic acid and ethylene glycol monomers that can undergo decarboxylation, esterification, and dehydration to benzoic acid, aromatic esters, and aliphatic hydrocarbon (Chen et al., 2019; Jin et al., 2021). The hydrocarbon distribution from mixed plastic exhibits an intermediate aromatic content (68.33%), which is lower than that in PS and PET but remains the dominant fraction (Figure 2), indicating significant contributions from the aromatic-rich PS and PET. The increased proportions of paraffins (11.61%) and olefins (7.59 wt%) are attributed to the presence of PP. Mixtures demonstrate synergistic effects, likely arising from radical mechanisms or the suppression of char formation due to mixed plastic interactions. The slight reduction in total aromatics indicates potential synergy toward aliphatic hydrocarbon formation, possibly facilitated by hydrogen transfer from PP to aromatic radicals (Boel et al., 2024; Ciuffi et al., 2021).

Conclusion

Non-catalytic hydrothermal conversion of individual polypropylene, polystyrene, polyethylene terephthalate and their mixture has been investigated with a focus on the oil products. The reaction temperature of 400 °C led to significant conversion of plastics in a 2 h reaction time. Polypropylene and polystyrene produced a mainly oil product, 73.65 wt.%, and 83.58 wt.%, respectively, while polyethylene terephthalate produced a significant amount of hydrochar (47.28 wt.%) as a solid residue. The mixed plastics provide an average oil (66.05 wt.%), gas (18.24 wt.%) and hydrochar (15.71 wt.%) yields due to synergistic interactions. The results showed evidence of synergistic interactions of the degradation products of the plastic feedstocks, leading to the production of deoxygenated oils dominated by fuel-range hydrocarbons.

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