

Inductively Heated Plastic Pyrolysis over H-ZSM5 Zeolite and Fluid Catalytic Cracking Catalyst

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The shift from a linear to a circular economy is an important step in combating global plastic pollution. Pyrolysis is a vital component of the circular economy, transforming unrecyclable plastic waste into valuable resources. However, plastic pyrolysis is an endothermic process with technical challenges due to its high energy demand that could potentially reduce its economic viability. To address this challenge, this study investigates the inductively heated thermal and catalytic pyrolysis of virgin plastics like high density polyethylene (HDPE), polypropylene (PP), polystyrene (PS) pellets and postconsumer plastic waste (PW) in a stainless-steel fixed bed reactor at 400-700°C. HZSM-5 zeolite and fluid catalytic cracking catalysts were employed as catalysts. All virgin plastics underwent complete plastic conversion, with PE and PP producing heavy hydrocarbons and PS generating high styrene yields through thermal pyrolysis. Catalytic pyrolysis over ZSM-5 catalyst notably increased gas yields for PE and PP due to enhanced cracking of pyrolysis intermediates. ZSM-5 significantly altered the liquid composition of PS pyrolysis by hydrogenating and isomerizing aliphatic chains on styrene molecules, despite having a minor impact on liquid yield. Despite the high char yield (25%) from PW, the permanent gases and gasoline exceeded virgin plastics due to the fluffy nature of PW samples, which enhanced transfer phenomena during pyrolysis. This study provides comprehensive mass balances for both thermal and catalytic pyrolysis of virgin and waste plastics, offering further insights into the role of pyrolysis in achieving a net-zero circular plastics economy.

1. Introduction

Global plastic production surged from 2 M t in 1950 to 380 M t in 2018, driven by population growth, lifestyle changes, and shale gas boom. In a business-as-usual scenario, the production of plastics is expected to reach 1,600 M t by 2050 (Rafey and Siddiqui, 2021). PE is the most abundant plastics used to date, followed by PP and PS (Ainali et al., 2021). The current global plastic production rate has far-reaching long-term and large-scale impacts that exceed the global capacity for assessment and monitoring. Given this dynamic, Persson et al. (2022) posits that plastic and other fossil-based chemicals need to be recognized as the tenth planetary boundary. Scientists, policymakers, and governments worldwide can combat plastic pollution by transitioning to a net-zero circular economy of plastics, aligning with Sustainable Development Goals (SYSTEMIQ, 2022).

The proposed new economy must focus on the decarbonisation of the energy supply and the substitution of fossil feedstock for circular carbon feedstock. In the European region, multiple driving forces exist for the transition to the net-zero circular economy of plastics, including (1) perspective change in the waste management sector, where plastic waste is regarded as feedstock for new products instead of waste to be

disposed of (Palos et al., 2021); (2) commitment at the regional level to achieve a climate-neutral economy and society by 2050, through European Green Deal and European Climate Law (Cifuentes-Faura, 2022), and (3) the urgency to reduce/eliminate further environmental pollution by microplastics, which carry persistent contaminants from the oceans to the global food webs (Wong et al., 2020).

As a complementary solution to mechanical recycling, chemical recycling is regarded as a vital strategy in the new economy to convert unrecyclable plastic waste into chemical feedstock (SYSTEMIQ, 2022). The global chemical recycling industry is expected to extract circular carbons from 100-210 M t of plastic by 2060 (Sederel et al., 2023). In 2022, the global chemical recycling capacity was approximately 1.2 M t. Significant development and investments are required to meet more ambitious targets. The Netherlands is among several leading nations actively developing chemical recycling technologies to meet national targets (Sederel et al., 2023), with an expected ~0.9 M t of plastic waste by 2027.

Pyrolysis is regarded as the most promising chemical recycling technologies by industry and policymakers. The technology is capable of converting mixed plastic waste feedstock (along with contaminants) into food-grade plastics production. In addition to the driving above forces, the long-term development of plastic pyrolysis technology is also favourable due to the following concepts: (a) a transition to a green chemical industry, where circular carbon feedstocks (including captured CO₂, recycled plastics, and sustainable biomass) and renewable electricity replace the roles of fossil fuels as carbon feedstock and energy source for chemicals production (Gabrielli et al., 2023; Lopez et al., 2023); (b) integration of plastic pyrolysis unit in the existing petroleum refinery system, due to the similarity in compositions of plastic pyrolysis products to crude oil and natural gas (Palos et al., 2021). From an industry perspective, such integration presents two major benefits: reduction of the high CAPEX cost related to upgrading and separation of plastic pyrolysis products for downstream applications, together with a shift of carbon feedstock from fossil fuels to circular carbon feedstock.

The conversion of polyolefin waste into light olefins is increasingly popular due to its alignment with the circular economy. Valizadeh et al. (2024) investigated converting waste plastic straw into valuable light olefins and monocyclic aromatic hydrocarbons (MAHs) via catalytic pyrolysis, finding that HZSM-5 (SiO₂/Al₂O₃:200) outperformed Hbeta (300) due to higher acidity and shape selectivity. Adding Co to HZSM-5 enhanced MAH production and propene selectivity. Netsch et al. (2023) studied catalytic pyrolysis of polyolefin-rich waste using zeolites and an amorphous silica-alumina catalyst in an auger-type reactor at 400–550°C, achieving high gaseous product yields (85 wt%) and significant C₂–C₄ olefin content (up to 67 wt%). This process also showed reduced coke formation (<3 wt%) and effective catalyst regeneration, though waste with nitrogen and halogen contaminants is unsuitable. These findings highlight the potential for efficient chemical recycling of polyolefin-rich waste into high-value base chemicals.

Despite the important roles of plastic pyrolysis technology, the high temperature required for the process leads to more significant climate impacts when compared to solvolysis (another chemical recycling technology) and mechanical recycling (SYSTEMIQ, 2022). Nevertheless, this technology could be an interesting opportunity to reduce the carbon footprint related to industrial production of chemical building blocks, especially C₂–C₄ hydrocarbons in petroleum refineries. The decarbonization of the existing chemical industry presents a significant challenge, as fossil fuels are used as energy sources and material feedstock. A life cycle assessment by Layritz et al. (2021) demonstrated that electrification of a traditional steam cracker only reduces ethylene production emission by ~30 %. The coupling of carbon capture and utilization technology to the electrified steam cracker could enhance net-zero ethylene production after 2035. This novel strategy is not cost-competitive in the near- and mid-term future. A more realistic scenario, as suggested by the authors, lies in the application of a (less energy-intensive) pyrolysis process and waste recycling strategy. This viewpoint is also supported by Mynko et al. (2023) in a separate analysis of the climate impacts produced by the global petrochemical industry. Plastic pyrolysis powered by induction heating at a laboratory scale has an energy saving of 80 % compared to plastic pyrolysis using an electric furnace (Wong et al., 2023). Research findings also demonstrate a high yield of gas products from the process due to the short residence time of plastic pyrolysis intermediates in the reaction zone, which minimizes secondary reactions. Such findings validate the potential of plastic waste as circular carbon feedstock for new chemical products including plastics. To further validate the potential of such technology in the net-zero circular economy of plastics, investigations on the pyrolysis of postconsumer plastic waste are necessary. Therefore, this study reports on the pyrolysis of virgin polymers, including PE, PP, PS, and real plastic waste collected in Madrid (Spain) under non-isothermal conditions. The complete mass balances for thermal and catalytic pyrolysis of the plastic samples are reported based on the full characterization of gas and liquid products, together with coke on catalysts. This study demonstrates the feasibility of postconsumer plastic waste conversion into light hydrocarbon gases (especially C₂–C₄ compounds) and gasoline, which can be used as feedstock for new plastic production. The research findings can potentially be extended to the pyrolysis of microplastics, which are abundant in PE, PP and PS (Shi et al., 2022).

2. Materials and Methods

Virgin polystyrene (PS), high density polyethylene (HDPE) and polypropylene (PP) pellets were obtained from a Spanish petrochemical company and used as received. A local municipal solid waste facility in Madrid (Spain) provided a plastic sample. The sample was dried and ground into small pieces of 0.5-1.0 mm. For catalytic pyrolysis, ZSM-5 catalyst (CBV 2314, Zeolyst International, $\text{SiO}_2/\text{Al}_2\text{O}_3 = 23$) and spent fluid catalytic cracking catalyst (E-Cat) obtained from a Spanish-based international energy company were used. Carbon disulphide (CS_2 , purity: $\geq 99\%$) was purchased from Pelkin Elmer and used as received. Extensive characterisations of the plastic samples and catalysts were previously reported by Wong et al. (2023). ZSM-5 were obtained in ammonium form after calcination at $550\text{ }^\circ\text{C} \times 5\text{ h}$. E-Cat was calcined at $700\text{ }^\circ\text{C}$ in the air for 3 h prior to the experiments to remove all the adhered impurities, moisture, and coke deposited under the conventional operation unit. This calcination step also transformed the zeolite from its ammonium form to hydrogen form. The catalytic pyrolysis of the plastics was performed in an inductively fixed bed reactor described in (Wong et al., 2022), which is connected to a gas product collection and analysis system. For each pyrolysis experiment, 2.0 g of plastic pellets (HDPE, PP, or PS) or shredded PW, together with 0.20 g of catalyst were examined under non-isothermal conditions. All the plastic samples (and the catalyst for catalytic pyrolysis) were placed inside the reactor before heating commenced. Prior to each pyrolysis process, the reactor system was purged with nitrogen gas (120 mL/min) for 10 min to ensure an inert environment. Next, the nitrogen gas flow was turned off. The induction heater was turned on for the specific period to allow reactor heating and the pyrolysis reaction. The heating rate was $70\text{ }^\circ\text{C/min}$ with a maximum temperature of $650\text{ }^\circ\text{C}$, which is consistent with previous conditions in the literature (Wong et al., 2023). The residence time of the plastic pyrolysis intermediates in the reactor was between 2.0 and 4.0 s. Under these reaction conditions, the plastic samples were converted to gaseous pyrolysis intermediates that exited the reactor based on the pressure difference with the external environment. Upon completion of the reaction, the reactor was allowed to cool to room temperature, and the condensed liquid product was removed and weighed. All the pyrolysis experiments were performed in duplicates, and average values were taken as representative values.

The amount of coke formed on the catalyst was characterised using the temperature-programmed oxidation technique (Xu et al., 2020). The light hydrocarbon compounds in the gas products were analysed using Agilent 8890 GC Systems based on established ASTM D5154 protocols. The boiling point distributions of the wax/liquid products were analysed according to ASTM method D2887-16 to determine the percentage of different fractions, namely naphtha ($<130\text{ }^\circ\text{C}$), kerosene ($130\text{-}216\text{ }^\circ\text{C}$), light cycle oil (LCO, $216\text{-}359\text{ }^\circ\text{C}$) and heavy cycle oil (HCO, $>359\text{ }^\circ\text{C}$). Simulated distillation of the liquid products was performed using a gas chromatograph (6890N, Agilent Technologies) installed with a Folio capillary column (ACCEL/CNS, 10 m, 0.53 mm, $0.88\text{ }\mu\text{m}$). The GC oven temperature was ramped from $40\text{ }^\circ\text{C}$ to $350\text{ }^\circ\text{C}$ at $35\text{ }^\circ\text{C/min}$, then held for 1.14 minutes. The chromatograph was equipped with an FID detector ($350\text{ }^\circ\text{C}$). Prior to analysis, the liquid sample was diluted with CS_2 (1:1 volume ratio). Mass balance was conducted for each experiment based on analysis results for gas, and liquid/wax products, together with coke.

Detailed hydrocarbon analysis (DHA) was also performed on the wax/liquid samples according to ATSM D6729 (Dunkle et al., 2019). An Agilent 7890A GC equipped with an Agilent 7693 autosampler was coupled to an FID for detection purposes. Instrument control and data processing were performed using Atlas 8.3 (V.9.00.0.10711, Thermo Fisher Scientific, Brede, the Netherlands). The polyaromatic hydrocarbons (PAH) compounds present in the products were analyzed by HPLC-RID (UV) Agilent and quantified using the standard EN12916 method. The analysis revealed a relative abundance of monoaromatics (MAH), di-aromatics (DAH), tri-aromatics (TAH), and polyaromatics (PAH) in the analyzed samples.

3. Results and Discussions

3.1 Thermal and catalytic pyrolysis of HDPE and PP

Complete plastic conversion was observed in HDPE and PP pyrolysis, indicating the effectiveness of the inductively heated fixed bed reactor in decomposing the plastics into lighter hydrocarbons. Thermal pyrolysis produced white or yellowish waxes, which was also observed by Wong et al. (2023). Table 1 displays the yields of different components in the plastic pyrolysis products, where gas product yield equals percentages of H_2 and C1-C4 hydrocarbons, and liquid product yield equals percentages of gasoline, LCO and HCO. As shown in Table 1, the reactions produced high yields of large hydrocarbons (C22+) but small amounts of gas products, signifying a low cracking degree. Extensive research on plastic pyrolysis shows that solid plastic undergoes melting and vaporisation to form intermediates in a gas state and experiences C-C bond scissions along the polymer chain via the free radical mechanism. In this study, a large volume of volatiles produced during pyrolysis exited the reactor rapidly. This resulted in limited heat transfer to the polymer chains and short residence time in the heating zone, which led to a limited degree of cracking. The slightly higher yields of C1-C12 hydrocarbons

from the thermal pyrolysis of PP (compared to PE) are related to the ease of C-C bond scission in PP due to the presence of tertiary carbons (Costa et al., 2022).

The catalytic pyrolysis of HDPE and PP over ZSM-5 zeolite resulted in significant cracking of heavy hydrocarbons (C13 and above) into lighter hydrocarbons. For HDPE pyrolysis, the yields of C3 and C4 compounds, and the liquid products in the gasoline range increased from 0.98 %, 0.66 % and 9.22 % (for thermal run) to 20.45 %, 21.20 % and 35.55 % (for ZSM-5). In comparison, the yield increases were less significant for PP, i.e., from 2.99 %, 0.44 % and 15.21 % (for a thermal run) to 11.30 %, 7.26 %, and 20.76 % (for a catalytic run). A similar observation was made by Wong et al. (2023). This is attributed to the different backbone structures of both plastics. The accessibility of pyrolysis intermediates to the catalyst particle's acid sites on its surface and in its pore, system determines the degree of cracking. For PP, the presence of methyl groups in the polymer chain backbone hinders diffusion of pyrolysis intermediates into the catalyst pore system and subsequent cracking steps on the active sites, leading to a lower cracking degree compared to PE.

Table 1: Product yields for thermal (denoted T) and catalytic pyrolysis over ZSM5 (Z) and E-cat (E)

Component	PE(T)	PE(Z)	PP(T)	PP(Z)	PS(T)	PS(Z)	PS(E)	PW(T)	PW(Z)	PW(E)
H2	0.00	0.10	0.02	0.08	0.00	0.05	0.05	0.89	1.90	0.83
C1	0.10	0.36	0.18	0.47	0.01	0.56	0.31	9.11	13.85	9.13
C2	0.59	3.35	0.59	2.39	0.03	1.05	0.71	15.50	15.08	13.28
C3	0.98	20.45	2.99	11.30	0.29	1.98	0.71	10.30	5.37	9.04
C4	0.66	21.20	0.44	7.26	0.05	1.21	0.35	3.60	3.12	5.55
Gasoline (C5-C12)	9.22	35.55	15.21	20.76	60.66	66.70	68.10	26.04	27.54	28.14
LCO (C13-C21)	20.52	8.77	14.91	12.81	21.75	14.65	15.06	9.42	4.45	6.02
HCO and wax (C22+)	67.92	9.01	65.66	44.23	17.21	11.93	12.28	4.94	2.12	3.11
Coke	0.00	1.22	0.00	0.70	0.00	1.21	2.44	0.00	4.21	3.14
Char	0.00	0.00	0.00	0.00	0.00	0.00	0.00	25.50	26.50	25.00

The yields from the catalytic pyrolysis products of HDPE and PP over E-Cat did not show significant improvements compared to thermal runs. The superior catalytic performances of ZSM-5 zeolite are related to its high porosity and acidity (Wong et al., 2023), which promote plastic depolymerization via the carbonium ion mechanism. As such reactions have been extensively studied and reported in the literature (Wong et al., 2023), more attention is paid to PS and PW pyrolysis in the following sections.

3.2 Thermal and catalytic pyrolysis of PS

PS, as a vinyl polymer, demonstrated extensive depolymerisation to form liquid products with a dark brown colour, even without a catalyst. The high yields of liquid products in the ranges of gasoline (60.66 %) and LCO (21.75 %) in Table 1 are related to the degradation of PS to oligomers. The different degradation behaviour of PS relative to HDPE and PP is attributed to its polymeric structure (with a phenyl group attached to every other carbon atom in the polymer chain backbone). Lu et al. (2021) proposed that PS degrades via a monomer dissociation mechanism, where the stereo-hindrance effect exerted by benzene rings in the polystyrene chain promotes scissions of bonding between a carbon atom of a styrene unit to the carbon atom adjacent to it, along the carbon backbone. Reaction parameters and catalyst properties can affect the degree of C-C bond scission (or depolymerisation) in PS, which in turn affects the abundance of monomers, dimers, and trimers in the liquid products. To further investigate the liquid product compositions, PAH analysis and DHA were performed.

As shown in Tables 2 and 3, 54.34 % of the aromatics in liquid products from thermal pyrolysis of PS was MAH, consisting mainly of styrene and minorly toluene. Catalytic pyrolysis over ZSM-5 significantly increased the relative abundance of MAH at the expense of DAH, TAH, and PAH (Table 2). This was accompanied by higher percentages of benzene, toluene and ethylbenzene and a lower percentage of styrene (Table 3).

Table 2: PAH analysis results on liquid products from catalytic pyrolysis of PS

Component	PS(T)	PS(Z)	PS(E)
MAH (% m/m)	54.34	59.70	58.64
DAH (% m/m)	10.46	12.08	9.19
TAH (% m/m)	12.37	8.07	11.49
PAH (% m/m)	22.83	20.15	20.68
Total aromatics (% m/m)	100.00	100.00	100.00

These results clearly indicate that ZSM-5 zeolite not only promoted the scission of C-C bonding along the polymer backbone but also the isomerisation and hydrogenation of aliphatic chains. Catalytic pyrolysis over E-

Cat also increased the percentages of MAH (although to a lesser degree than ZSM-5), which contained more benzene, toluene, and ethylbenzene compared to the thermal runs. Such results indicate that E-Cat could also promote PS degradation to styrene, in addition to its hydrogenation to ethylbenzene.

Table 3: Mass percentages of major compounds in pyrolysis liquid products of PS over different catalysts according to DHA results

Compound	PS(T)	PS(Z)	PS(E)
Benzene	0.07	17.04	1.89
Toluene	1.09	5.63	1.89
Ethylbenzene	0.14	2.67	5.28
Styrene	58.99	27.41	48.75
Xylene	0.00	0.55	0.00
C10 Aromatics	0.00	0.09	0.69
Total	60.28	53.39	58.50

3.3 Thermal and catalytic pyrolysis of real plastic waste

Thermal and catalytic pyrolysis of PW produced nearly 25.0 % of char (Table 1), signifying the presence of inert materials and possibly biomass materials that could not be pyrolyzed under the experiment conditions. However, thermal pyrolysis of PW produced higher yields of C1-C4 compounds than all the virgin plastics tested, especially for methane (9.2 %), ethane (13.3 %), propylene (11.0 %), and ethylene (3.7 %). These observations are most probably related to the different shapes of plastic samples (fluffy for PW and granular for HDPE, PP, and PS pellets). Jeon et al. (2021) investigated the pyrolysis of fluffy and bundled plastic packaging waste in a semi-batch reactor, which produced distinctive product yields and distribution. They reasoned that the shapes of the plastic feed affected heat transfer to the polymeric molecules and also the residence time of the pyrolysis intermediates in the reactor. In their work, C-C scissions in plastic flakes were accelerated due to high heat transfer rates from the reactor environment to the polymeric chains, resulting in more rapid evolutions of C1-C4 hydrocarbons. On the other hand, bundled plastics were pyrolyzed at a slower rate (due to a lower heat transfer rate to the whole plastic sample), which produced a higher yield of gasoline fraction. In this study, the volatile intermediates exhibited a very short residence time in the fixed bed reactor. This occurrence minimized the occurrences of fragments recombination into larger hydrocarbons, which resulted in lower yields of liquid products (C5 and above). Another possible explanation is the potential presence of organic materials and metals in PW that led to co-pyrolysis with enhancement of catalytic effects. The ZSM5 zeolite can crack larger hydrocarbons, leading to higher yields of H₂ and C1-C2 hydrocarbons and lower yields of hydrocarbons with carbon numbers of C13 and above. In comparison, E-Cat marginally increased the yields of gasoline and C4 hydrocarbons compared to thermal runs. These observations are consistent with the results described in Sections 3.1 and 3.2.

4. Conclusion

Inductively heated pyrolysis of virgin and postconsumer plastic materials was performed in a fixed bed reactor to reveal its product yields and compositions. Thermal pyrolysis of HDPE and PP produced high yields of heavy hydrocarbons but small amounts of gas products, while PS produced high yields of monomers in addition to dimers and trimers. This difference is attributed to the distinctive polymer chain structures of the plastics, which led to depolymerisation via different mechanisms. PW, albeit with a high char yield, demonstrated high yields of permanent gases, most probably due to the fluffy shapes, which accelerated degradation and cracking. Catalytic pyrolysis over ZSM-5 significantly increased yields of permanent gases and gasoline for all plastic samples tested, evidenced by a higher degree of cracking on the catalyst acidic sites. E-Cat demonstrated similar catalytic effects to those of ZSM-5 zeolite to a lesser degree. This study indicates that plastic waste is a possible source of C2 and C3 hydrocarbons, as well as gasoline-like hydrocarbons, which could be used as feedstock for new plastic materials and other value-added products.

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