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Original Article

Product Development from Waste Polymer Packages Through Pyrolysis

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Abstract - *Pyrolysis of waste plastics is a novel method for effective waste management. Waste polyethylene, polypropylene, and their mixture were pyrolyzed in a tubular batch reactor at about 300-350 °C in the presence and absence of a catalyst. Polyethylene and polypropylene were chosen because they are the most abundant waste polymers. The polymers decomposed into a gas, a liquid, and a solid. The liquid product was further distilled to obtain fractions with properties and compositions similar to those of gasoline and diesel. Innumerable reactions occur during the pyrolysis of plastics, with distinct kinetic parameters. Various tests, including density, aniline point, flash and fire points, and GC-MS analysis, were performed to characterize them.*

Keywords - *Polymers, Polyethylene, Polypropylene, Diesel, Gasoline.*

1. Introduction

The major components of waste plastics are polyethylene and polypropylene, as they are widely used in packaging and are generally discarded after a single use. Only a small fraction of these tremendous amounts of waste plastic is being recycled/reused; most of it is either landfilled or thrown around us. Another problem arising from the haphazard disposal of plastics is the clogging of the sewage system, as the waste materials ultimately flow into it. Moreover, plastics are not biodegradable. However, some varieties may degrade after a long exposure to sunlight. The disposal of large amounts of plastic has led to complex waste management systems. Various methods are available for the disposal of plastic waste, such as landfilling, incineration, and pyrolysis. Amongst these, pyrolysis is one of the most efficient processes, as it does not produce any obnoxious gases. The present rate of economic growth would be unimaginable without the conservation of fossil fuels, such as crude oil, natural gas, and coal; however, the manufacturing of plastics consumes a considerable amount of petroleum.

Classically speaking, there are three principal modes of managing this waste stream known globally, viz.

- Using thermosetting plastics (waste) as a carbon source for the carbon reduction process in metallurgical operations.
- To produce petroleum feed stocks by the catalytic degradation of the thermosetting plastics, and
- To use as a source of energy by mixing it with fuels in the combustion chamber.

Pyrolysis is the thermochemical decomposition of organic material at elevated temperatures in the absence of oxygen. Further degradation of these subsequent molecules depends on several factors, including (but not limited to) temperature, residence time, catalysts, and other process conditions [1─4]. The pyrolysis reaction can be carried out with [5─11] or without [1─3; 12─15] the presence of a catalyst and in different types of reactors, namely batch [3; 12; 16─18], semi batch [11; 19], and continuous, like fluidized bed [5; 7] and autoclaves [3; 8] etc. In another continuous reactor, plastic in the form of a paste is fed into a heated column using a screw conveyor [3; 9; 14]. The products obtained during the process are a solid residue (mainly carbon in the form of soot), a liquid (a mixture of hydrocarbons), and a gas [20; 21]. The residue contains mainly carbon, with occasional ash [3]. Different types of catalysts, acid silica–alumina [20] or zeolite (HY, HZSM-5, mordenite) [4; 21─24], cracking catalysts [7; 9], alkaline compounds like ZnO, CaO, and K2O [18] were used by different researchers.

A few researchers also pyrolyzed polymers mixed with petroleum- or carbon-derived fractions in the presence of cracking or hydrocracking catalysts [23]. The extent of conversion of plastics or plastic-derived waxes into light engine fuels can be enhanced using stable hydrocracking catalysts.

Temperature and residence time play important roles in the pyrolysis of plastic materials. Innumerable reactions with

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different orders take place during the operation. The formation rates of different products depend on operating conditions such as temperature, residence time, and catalysts [26]. Thermogravimetric studies provide data on the gradual degradation of polymers as temperature increases and are among the methods used to determine the reaction kinetics of pyrolysis.

Pyrolysis experiments on Refuse Plastic Fuel (RPF) were performed in a tube furnace under non-isothermal conditions [26] in a nitrogen atmosphere, and the authors also discussed the effect of temperature on the yields of liquid, solid, and gaseous products. Thermal decomposition characteristics of Polystyrene (PS), Polyvinyl Chloride (PVC), etc., along with RPF during pyrolysis were found using a thermogravimetric analysis reactor. Using the single-reaction model, they determined the activation energy and pre-exponential factor for RPF pyrolysis and used the parallel-reaction model to determine the activation energies for the cracking of PS, PP, LDPE, PVC, etc.

The pyrolysis of plastic, tire, coal, etc., at different heating rates was studied by some researchers [27], who examined the behavior of different samples under non-isothermal pyrolysis conditions using the distributed energy model.

The pyrolysis behavior of different polymer samples, such as HDPE, LDPE, PP, PS, ABS, and PET, individually or as mixtures of some of them, was observed in different reactors [28; 29] isothermally or non-isothermally. Kinetic models were developed using thermogravimetric studies of some polymers [28; 30─34]. Whenever a polymeric mixture was degraded, the overall degradation rate, which correlated with the degradation rates of the different pure components, was determined [33]. Some researchers determined the activation energies for the decomposition of different polymers using a first-order kinetic model [34]. They also concluded that the activation energies of thermal decomposition of the polymers decreased with increasing heating rates.

Pyrolysis of municipal wastes (mixture of PE, PP, PS, etc.) was carried out in a horizontal tubular reactor [35] and in a shaft kiln, where the temperature distributions of the gas and solid phases could be separately controlled [29]. A simulation model that reproduced the product distribution of gas-phase reactions of volatiles from biomass and polymers had been developed.

Thermal decomposition of commingled plastics comprising HDPE, LDPE, PP, and PVC under vacuum was investigated, and it was observed that interactions occurred during pyrolysis, mainly at high temperatures. The chlorine from PVC in the mixture was released almost completely before 375 °C [36].

The amount of tar decreased with increasing pyrolysis temperature during the pyrolysis of municipal waste [29]. The objectives of this project were to obtain substitute fuels from pyrolysis products and to observe the effects of catalysts, ZnO, and fly ash on the process.

2. Experimental

2.1. Material

Polyethylene and polypropylene pellets obtained from Haldia Petrochemicals Ltd, shredded waste polyethylene and polypropylene bags, collected from household wastes. Zinc oxide and fly ash were used as catalysts.

2.2. Experimental Setup

The experimental setup is shown schematically in Figure 1. The batch reactor, cylindrical in shape, was made of stainless steel and had a flange connection. It had an O.D. and I.D. of 0.055 m and 0.05 m, respectively, with a length of 0.45 m. The reactor had the outlet at the top, and there was an arrangement to flush the reactor interior with nitrogen. Thermocouples were placed at different locations as shown in the figure. The reactor could be heated using an electrically operated heating arrangement (capable of heating the reactor contents to 6000 °C) in the form of a cladding. Outside the cladding, there was a layer of insulation.

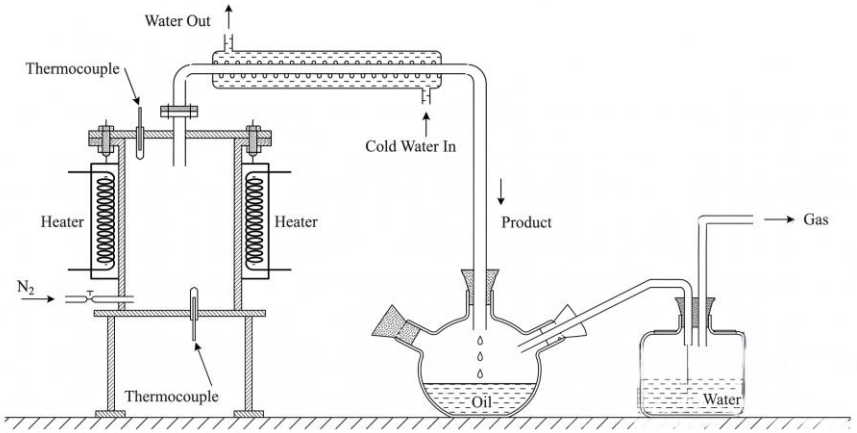


Fig. 1 Schematic diagram of the pyrolysis unit

The product gases leaving the top of the reactor were partially condensed in a condenser to separate the liquid and gas phases. The liquid product was collected in a closed container having an outlet for the gas. The gas evolved was flared after metering. Before metering, the gaseous product was passed through a bed of water so that no liquid product (tarry) could choke the gas meter. The liquid product obtained was further subjected to ASTM distillation, and the characteristics of the fractions were determined to assess their suitability for use as a substitute for gasoline or diesel. Boiling ranges of gasoline and diesel available in the market are 35-120 °C and 120-220 °C, respectively. The fractions obtained during ASTM distillation within these ranges were selected, and various tests, such as aniline point, flash point, fire point, and density determination, were performed on these fractions. Major components were identified by GC-MS.

Pure as well as waste Polyethylene (PE) and Polypropylene (PP) samples were pyrolyzed. Whenever waste polymers were used, the bags were shredded and subsequently pyrolyzed. In the polyethylene-polypropylene mixture, a 1:1 by weight ratio was maintained. Zinc oxide was used as a catalyst for pyrolyzing waste polyethylene and polypropylene, but for their mixture, zinc oxide, as well as fly ash, was used.

100 g of sample in each case was fed to the reactor, flushed with nitrogen, and pyrolyzed to 4000 °C. The product gases leaving the top of the reactor were partially condensed to obtain a liquid and a gas. The amount of gaseous material produced was approximately 5 liters in each case.

The liquid product was collected in a closed container having an outlet for the gas. The liquid product obtained after pyrolysis was fractionally distilled, and the fractions collected during distillation at three temperature ranges (up to 120 °C, 120-220 °C, and 220-260 °C) were collected. The boiling ranges of petrol and diesel available in the market are 35-120 °C and 120-220 °C, respectively. So the fractions obtained during ASTM distillation within these ranges were selected. The characteristics of the fractions obtained were assessed to determine their suitability as substitutes for gasoline or diesel. We could not distill beyond 300 °C because the glass apparatus used could fail. Above 220 °C, the product resembled lubricating oil and exhibited high viscosity. For waste polyethylene, there was no significant change in the previously mentioned data. However, the solid residue produced slightly more ash. The amount of gaseous material produced was approximately 5 liters in each case. Principal components were identified using a Thermo Scientific GC-MS. However, in both cases, the column was a DB-5MS column.

Pyrolysis of 100 g each of PE and PP, and their mixture, yielded approximately 50 mL of liquid product and 30 g of solid residue. Distilling the liquid product obtained during pyrolysis of the PE and PP mixture produced a 20% volume fraction with a boiling point up to 120 °C, a 35% volume fraction with a boiling point within the range of 120 to 220 °C, a 25% volume fraction with a boiling point within the range of 220 to 300 °C, and the rest above 300 °C. Above the temperature of 220oC the product was similar to lubricating oil having high viscosity. Above 300 °C, we could not proceed as the glass apparatus used could give way. The fire points of the different samples depended on the distillation temperature, not on the sample fed for pyrolysis. The fire points of the samples obtained from the two fractions, up to 120 °C and 120-220 °C, were 36 °C and 52 °C, respectively. The Aniline Points were found to be 36 °C and 65 °C, respectively. However, gas production was slightly higher whenever fly ash was used. Fly ash mainly consists of oxides of silicon and other metals.

Pyrolysis of PE, PP, and their mixture was also attempted at an elevated temperature of 800 °C. It was a quick heating process. The sample amount taken in each case was 500 g, and it was rapidly heated to the requisite temperature. The experiment yielded approximately 50 liters of gas and a meager amount of liquid product.

3. Results and Discussion

Table 1. Characteristics of different products obtained after pyrolysis of different samples of polyethylene

Sample no.	Raw materials	Amount of liquid product obtained (ml/100 g)	Residue C gms/100 gm of sample	%Ash
1	Pure polyethylene without a catalyst	101	11	0.2
2	Waste polyethylene without a catalyst	60.17	13	17.26
3	Waste polyethylene with a catalyst	60	14.7	19.74

Table 2. Characteristics of different products obtained after pyrolysis of different samples of polypropylene

Sample no.	Raw materials	%Amount of liquid product obtained (ml/100 g)	Residue C gms/100 gm of sample	%Ash
1	Pure polypropylene without a catalyst	95	8	4.54
2	Waste polypropylene without a catalyst	60.17	10	5.17
3	Waste polypropylene with a catalyst	64.9	14	6.5

Table 3. Characteristics of different products obtained after pyrolysis of different mixtures of polyethylene and polypropylene

Sample no.	Raw materials	%Amount of liquid product obtained (ml/100 g)	Residue C gms/100 gm of sample	% Ash
1	Waste PE and PP mixture	60	11	0.2
2	Waste PE and PP mixture with ZnO as a catalyst	61	13	17.26
3	Waste PE and PP mixture with fly ash as a catalyst	60	14	19.74

4. Discussion

Products were observed to form at nearly 275 °C, and the formation rate increased with temperature. The pyrolysis is a very slow process. Production was found to be maximum at 350 °C. The total time required for complete pyrolysis was approximately 2 hours. Slow heating is necessary for this process, as the liquid products are highly likely to crack into gaseous products. The product stopped flowing out at a temperature of about 430 °C. Another observed peculiarity was a reduction in both the time and the final temperature when using a trace amount of CaCO3 along with ZnO. However, there was no change in quality by using the second component.

Product components obtained have been observed to vary a lot depending on the type of raw material fed for pyrolysis. Catalyst also plays an important role in product formation and has been observed to alter the products. Compounds containing bromine were detected in the product stream. The reason is the coloring material present in the raw material (waste PE&PP), which produced those bromine-bearing compounds. A small amount of wax was also formed during the process. The residue was found to be char formed by the cracking of the oil-type fluid. The gases evolved may be used as a fuel.

While performing experiments with these three samples, pure PE, waste PE, and waste PE with ZnO, it was observed that whenever the amount of liquid product obtained was less, the amount of gas evolved was more. When pure polyethylene was pyrolyzed in the absence of a catalyst, the amount of liquid product obtained was much higher, but it contained more grease, as observed by ASTM distillation. Polyethylene pouches produced a satisfactory amount of liquid product, but the grease content was also quite high. Catalyst (ZnO) slightly improved the production rate. However, the quality of the finished product obtained was not very good. The aniline points of those fractions were determined and were observed to closely resemble those of gasoline and diesel. Other properties, such as flash point and fire point, did not vary much across all cases. It was found that the chemical structure of polymers significantly influenced the qualitative and quantitative properties of the volatile products.

The yield of volatile products increased both with temperature and residence time. The yield of liquids of 10–25% separated from the gases leaving the reactor could be increased by a further 10–15% by distilling the residue. After distillation, the aromatic content was concentrated primarily in the lighter fraction (F1), an advantageous property for further fuel-like utilization. As with the aromatic content, the heteroatom content of the product liquids was also influenced by the raw material's structure.

During the pyrolysis of polypropylene, the yield of volatile products increased with both temperature and residence time. The yield of liquid (10–25%) separated from the gas leaving the reactor could be increased by 10–15% by distilling the residue. After distillation, the aromatic content was concentrated primarily in the lighter fraction (F1), an advantageous property for further fuel-like utilization. As with the aromatic content, the heteroatom content of the product liquids was also influenced by the raw material's structure.

More than one hundred products of pyrolysis of waste PE& PP mixture with ZnO as catalyst were formed during fractionation of the middle fraction (boiling range 120 °C to 220 °C. However, fewer components were present in the products when the PE & PP mixture was pyrolyzed with fly ash as a catalyst.

More than 100 products of pyrolysis of a waste PE & PP mixture, with fly ash as a catalyst, were formed during fractionation of the middle fraction (boiling range 120 °C to 220 °C). The details of the product obtained during pyrolysis are shown in SL1 (Supplementary).

5. Conclusion

Pure polyethylene produced the highest-quality oil during pyrolysis; afterward, oil was also produced from waste polyethylene. The quality and quantity of oil produced during the pyrolysis of polypropylene came next. Whenever the mixture of polyethylene and polypropylene was pyrolyzed, the product should have been an average of PE and PP pyrolysis, but it did not turn out that way. ZnO, as a catalyst, performed well, as reported in the literature, but fly ash's performance is notable. However, the suitability of the final product is to be examined.

The plastic waste, if used properly, i.e., when pyrolyzed, can provide cheap fuel oil as a substitute for diesel. Best results are obtained by blending this oil with diesel. In another way, it will help dispose of a large amount of plastic waste. All the products, gaseous, liquid, and solid, are useful. So, by eliminating hazardous materials from the mixtures, we can definitely obtain valuable products through the pyrolysis of polymeric wastes.

Conflicts of Interest

Authors declared that they have no conflict of interest to disclose

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