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Catalytic Pyrolysis of Polyethylene Waste: A Comparative Study of Thermal and Catalytic Degradation

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Abstract

The increasing accumulation of plastic waste is one of the major concerns in the contemporary world. India's estimated daily production of plastic waste in the fiscal year 2021-22 is around 26000 tons. The pyrolysis provides methods for the removal of this waste in a constructive manner by transforming it into a usable fuel. In this research work, the catalytic and non-catalytic pyrolysis of polyethylene (PE) waste was studied using magnesium carbonate ($MgCO_3$) as a catalyst. The non-catalytic method yielded a wax fraction containing 60% (w/v) at 450°C while the catalytic method gave 70% (w/v) clear brown liquid fraction. The properties of the liquid fractions were compared with international diesel fuel, showing several similarities with the exception of flashpoint.

The GC-MS results indicate that the catalytic products had a liquid compound within the observable limits C_{10} to C_{15} which is comparable to diesel. The char from catalytic pyrolysis was better in terms of porosity revealed through SEM image analysis. XRD images confirmed that the activated char still contained some MgO catalyst, indicating some potential for catalyst recovery and reuse.

Keywords: Polyethylene, HDPE, LDPE, Plastic waste, Pyrolysis, Catalyst.

Introduction

According to data from the New Hampshire Department of Environmental Services, the world generated 242 million tons of waste in the form of plastic, which accounted for 12% of all municipal solid waste in 2016. As plastic waste accumulates, its visibility has increased, posing negative environmental and health impacts. Plastic garbage can take hundreds to thousands of years to break down in the environment, in contrast to organic waste[1]. The Central Pollution Control Board (CPCB) reported that India generated approximately 9.4 million tons of plastic waste per annum in 2021-22[2], which amounts to 26,000 tons of waste per day. Of this, 5.6 million tons per annum were recycled, while 3.8 million tons per annum were left uncollected or littered, amounting to 9,400 tons of waste per day[3]. Approximately 70% of plastic packaging products are converted into plastic waste in a relatively short period[4]. When plastic is thrown away after its useful life is ended, plastic waste is produced. India consumes approximately 11 kilograms of plastic per person on average, which is far less than the global average of 28 kg. But according to a Ministry of Petroleum and Natural Gas forecast from the Indian Government, the country's annual per capita consumption is expected to reach 20 kg by 2022[5]. Since pyrolysis converts waste into energy, it is an easy means to recycle used plastic.

Pyrolysis is the irreversible thermal degradation of complex long-chain organic polymer molecules into short-chain polymer that occurs endothermically at temperatures higher than 300°C in the absence of atmospheric oxygen, with or without pressure[6-8]. The primary products of the pyrolysis of plastics include gas, liquid and char, with liquid oil being the desired outcome[9,10]. The escalating depletion of renewable fossil fuels, which have a replenishment time of millions of years, is intensified by their growing demand in our growing world. This presents a substantial challenge for the future, worsened by the surging need for plastics, which are derived from petroleum-based resources. As a solution, converting plastic waste into fuel has been developed. Since plastics are derived from petroleum, the oil produced through the pyrolysis process has a high caloric value and can be used as an alternative fuel[11-13]. The primary obstacle in pyrolysis is the effective conversion of waste plastic into fuel while simultaneously minimizing energy usage to produce valuable commodities like gasoline or diesel. To overcome these constraints, it is essential to choose the right catalyst and compare it with thermal degradation[8,14,15].

Material and Methods

Polyethylene: For this experiment, we chose to use polyethylene plastic bags, specifically those made from a combination of HDPE and LDPE. Polyethylene is a type of plastic commonly used in the production of poly bags and plastic toys. It has a straightforward structure and offers a wide range of versatile applications Plastic waste from packaging and containers made of PE accounts for approximately 56% of all plastic waste, with 75% of it originating from residential households. By reducing the amount of PE used, we can minimize the amount of waste that ends up in landfills and avoid negative environmental impacts[16,17].

Catalyst: $MgCO_3$ is used as catalyst in the reaction. The primary products of plastic waste pyrolysis are carbon dioxide, methane, hydrogen and carbon monoxide[18].

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Calcinations of $MgCO_3$ to produce MgO in the presence of waste plastics has resulted in up to 99% carbon dioxide and methane reduction was demonstrated. The $MgCO_3$/MgO catalyst action in efficient carbon reforming reactions with diminished or no carbon deposition is demonstrated[19]. Crude oil yield was higher using $MgCO_3$ as catalyst. $MgCO_3$ is used as basic and easily available catalyst in catalytic degradation[20].

Flow chart: The experimental process is depicted in figure 1 as a scheme of pyrolysis. The plastic waste (PE) was cleaned and chopped before being added to the pyrolysis flask. In the process of thermal degradation, 50 grams of waste plastic were utilized, while an additional 50 grams of plastic waste were combined with 1 gram of $MgCO_3$, as a catalyst for the catalytic degradation process. Nitrogen gas was injected into the reactor to remove oxygen. The pyrolysis was carried out at a temperature of 450°C for 90 minutes in a pyrolysis apparatus that was placed inside a chemical fume hood to exhaust gases and vapors. The experimental procedure of pyrolysis necessitated the continuous circulation of water for the purpose of condensation; we have structured the system to ensure that water is circulated efficiently, thereby minimizing waste in the recycling process.

The fuel/wax fraction was collected and evaluated based on the physiochemical characteristics of petroleum fuel tests. The physical characteristics of the liquid samples that were collected were determined using standard ASTM procedures for fuel tests. A viscometer was used to measure the samples' viscosity, specific gravity was computed numerically and the samples' density was assessed using the standard IS 1448 Part 32 procedure. API gravity (ASTM-D1298-85) and kinematic viscosity (ASTMD445-87) were investigated ASTM methods. Flash and fire point were determined by open cup apparatus (ASTM-D92-78) and pour point was determined with the help of pour point apparatus. Calorific values of the samples were determined by using Rico Scientific Industries (Model: RSB 7.6) Bomb Calorimeter.

Remaining residue (char) in the flask is collected for further Proximate and Ultimate analysis. To obtain a powder with smaller particles, the char that was produced after pyrolysis of waste PE plastic was sieved through muslin cloth. Using ASTM Methods, a proximate examination of the char's moisture, volatile matter and ash contents was carried out. Calorific value was determined according to ASTM D 5865, using Rico Scientific Industries (Model: RSB 7.6) Bomb Calorimeter.

Activated Carbon Preparation: HDPE and LDPE plastic are composed of many materials such as polyethylene polymer which itself added with materials such as plasticizers, filler and dye (pigment) etc. to prepare activated carbon from char. Char was activated thermally with ammonium chloride in 1:2 proportion under air atmosphere at 900°C for 3 hours in Muffle furnace.

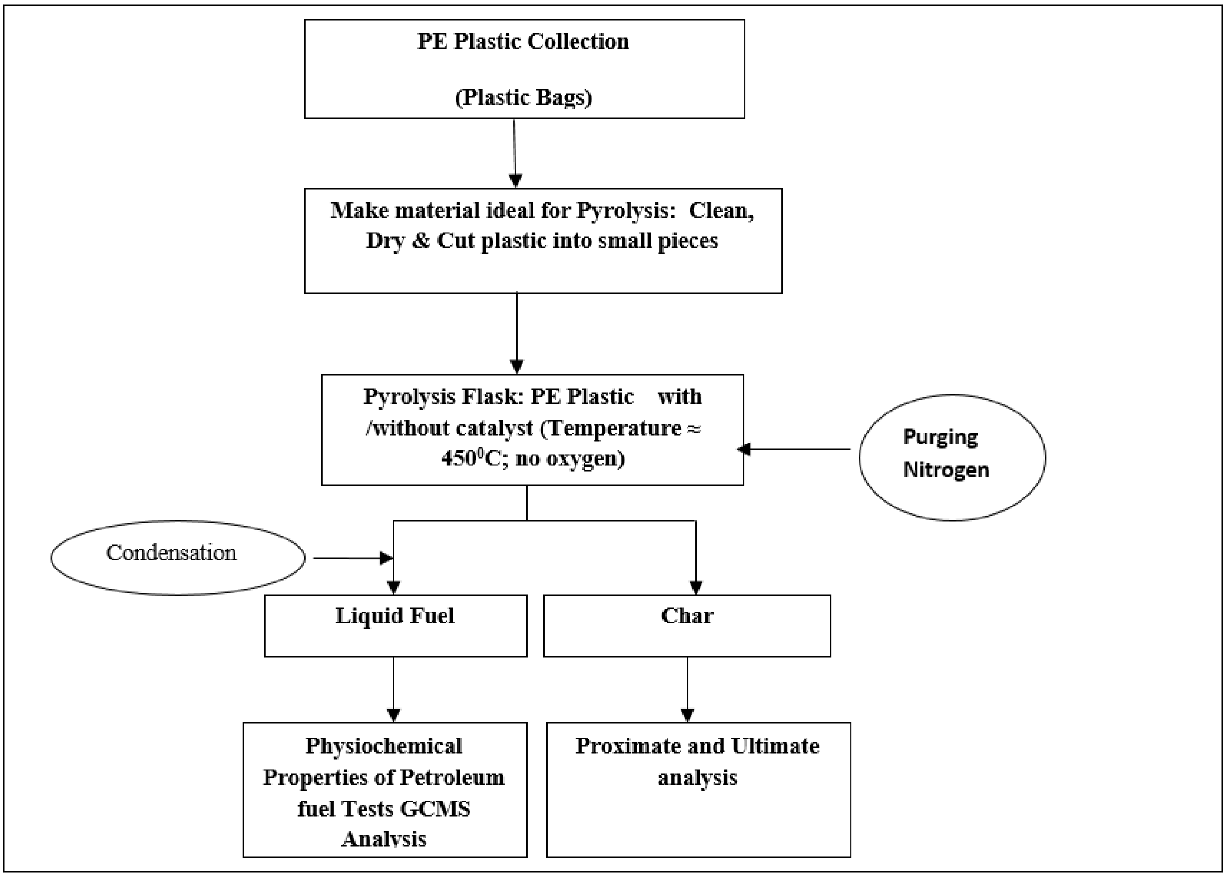

Figure 1: Experimental Scheme of Pyrolysis

Results and Discussion

Thermal Degradation: Thermal degradation of PE was carried out upto the temperature 450°C. Up to 260°C, no reaction was seen, above this temperature a slight conversion into wax and gases took place, as it was followed by a gradual rise in the liquid yield and the overall percent conversion as the temperature rose to 450°C. The total percent conversion was maximum at 450°C in 90 minutes. On thermal degradation, 30ml of wax fraction (60% w/v) was collected from 50 gm of PE waste.

Catalytic Pyrolysis: 50 grams of plastic waste along with 1 gm of $MgCO_3$ as catalyst for catalytic degradation are added into pyrolysis flask. Pyrolysis was carried out at 450°C for 90 minutes. On catalytic degradation with $MgCO_3$, 35 ml (70% w/v) of fuel fraction was collected which is clear brown colored liquid fraction. Using catalyst complete conversion into liquid yield with selectivity towards the liquid increases with decrease in wax production[20,21].

Different kinds of fuel tests for physical properties were used to determine whether the liquid produced by thermal and catalytic pyrolysis of PE was suitable as a fuel oil[22]. Table 1 provides a summary of the findings and for comparison, the standard values of standard diesel oils are used. Physical characteristics of thermally and catalytically produced liquids were measured, including density, specific gravity, viscosity, flash point and calorific values. When comparing the physical characteristics of the liquid samples to the usual physical test values of diesel, it was observed that the oil fraction exhibited identical physical characteristics, with the exception of flash point.

GC-MS Analysis: GC-MS analysis of liquid obtained after thermal and catalytic pyrolysis was done. Gas Chromatography shows presence of hydrocarbons in the range of $C_{12} – C_{29}$ with liquid obtained by thermal Pyrolysis while $C_{10} – C_{15}$ hydrocarbons range is observed with liquid obtained by pyrolysis with the use of catalyst. According to GC-MS results, we can predict that oil obtained using thermal pyrolysis is crude oil which needs further refining and the oil which is obtained using catalytical pyrolysis is with hydrocarbon range $C_{10} – C_{15}$ which shows similarity with diesel hydrocarbon range[23,24].

Table 1
Data of the physical parameters of thermal and catalytic derived liquid fractions with standard diesel oil

S.N.	Physical Parameters	Fraction on Thermal degradation	Fraction on Catalytic degradation	Standard Values of Diesel
1	Density Kg/L at 15.6°C	0.839	0.848	0.82 - 0.88
2	Specific Gravity	0.873	0.879	0.82 - 0.88
3	Viscosity centipoises	4.05	3.00	2 – 4.5
4	Flash Point	49°C	44°C	55°C - 66°C
5	Calorific Value cal/g	45.3	43.9	43.7

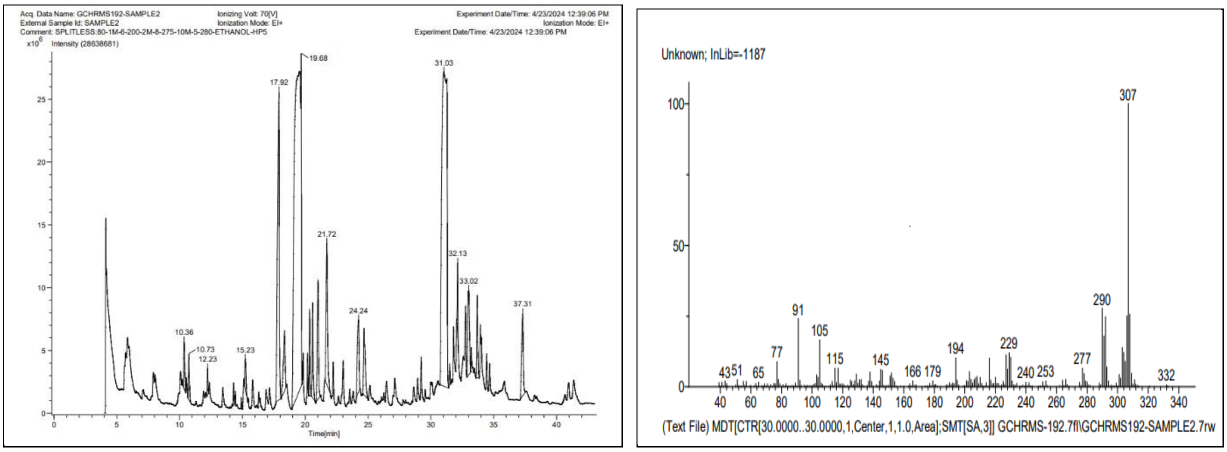

Fig. 2: Gas Chromatogram and Mass Spectra of liquid Fuel without catalyst

Table 2
Characteristics of chars derived from thermal and Catalytic Pyrolysis.

Test	Parameters	Char after Thermal Pyrolysis	Char after Catalytic Pyrolysis
Proximate Analysis (% Wt)	Moisture	1.93	2.05
	Ash	0.18	0.55
	Volatile Matter	56	53.5
	Fixed Carbon	41.89	43.9
Partial size (g/Cm^3)		0.378	0.0491
Calorific Value cal/g		3900	4500

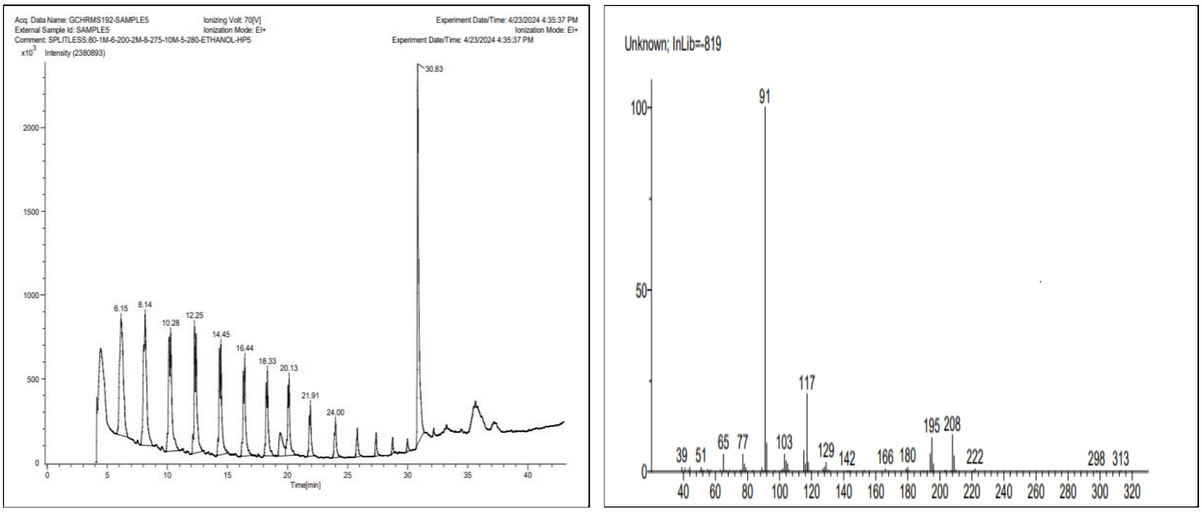

Fig. 3: Gas Chromatogram and Mass Spectra of liquid Fuel with catalyst $MgCO_3$

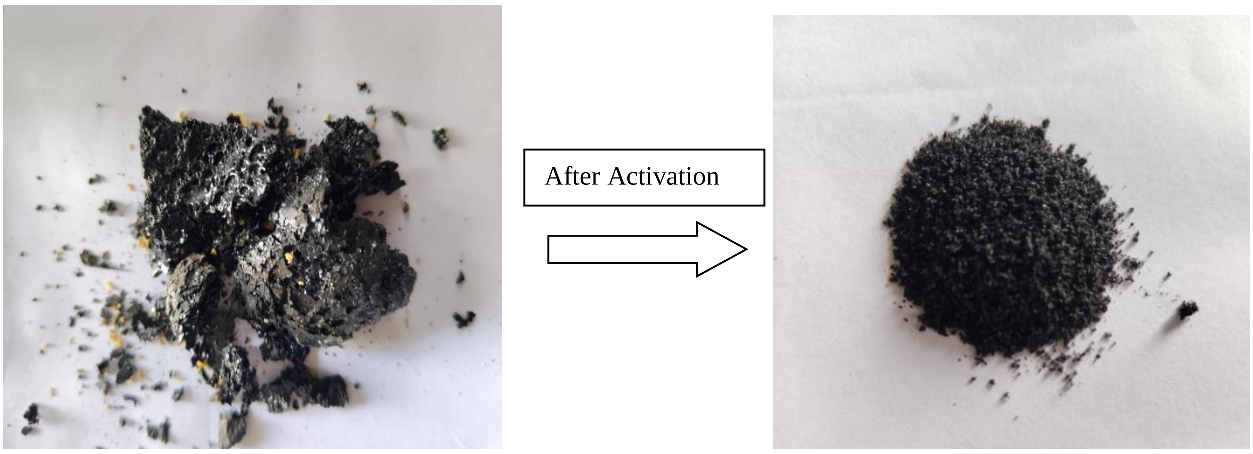

Fig. 4: Char remaining after catalytical pyrolysis **Activated carbon**

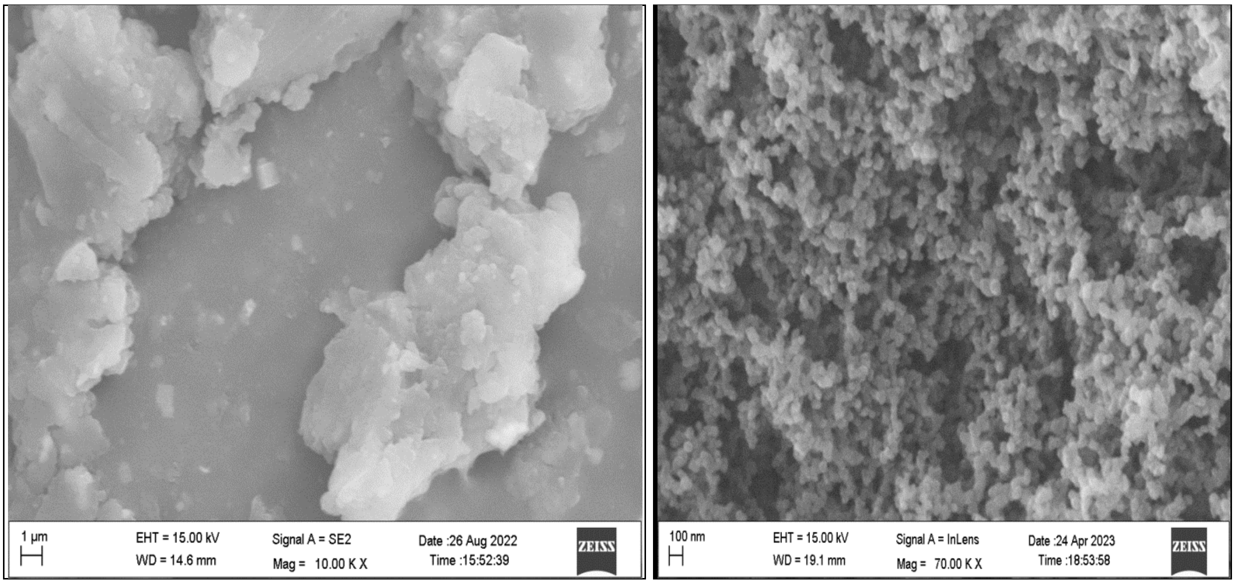

Fig. 5: SEM Image of char derived from catalytic pyrolysis of plastic wastes
(a) without activation; (b) with activation under air atmosphere at 500°C for 3 hours.

Characterization of char derived from thermal and Catalytic Pyrolysis: Proximate analysis, density and calorific value of the char remain after thermal and catalytic pyrolysis of PE wastes was summarized in table 2. The volatile matter and fixed carbon must be the main components because plastic was generally composed of 75 - 100 % volatile matter, 0.1- 2.75% moisture content and 0.2 - 4% ash.

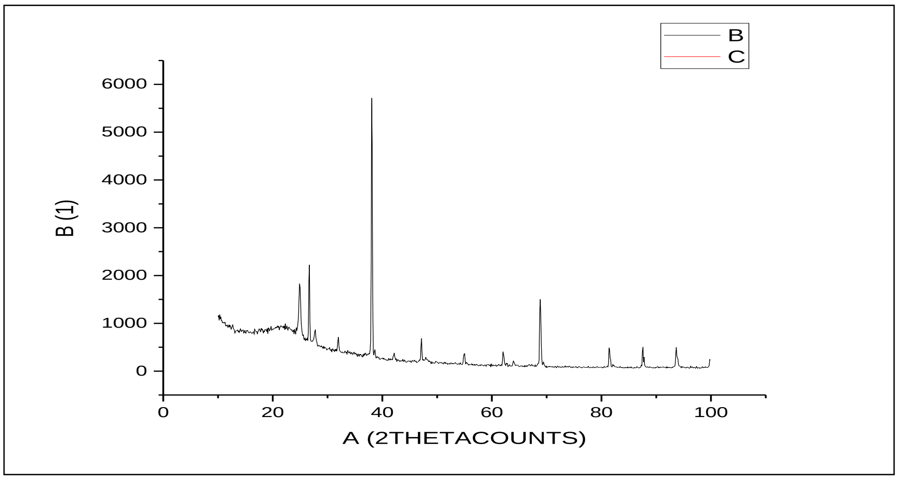

Fig. 6: X Ray Diffraction Pattern of MgO seen in activated char obtained after catalytic pyrolysis

After being thermally decomposed via pyrolysis process, volatile matter was still in majority whereas moisture and ash were in minority[25]. Additionally, table 2 showed that the calorific values of the char produced by both thermochemical procedures were lower than those of the common polymers.

Morphology and Pore Properties of obtained Char and Activated Char: The importance of studying activated carbon, as the primary carbon component in char, is significant. The morphological characteristics of the char obtained from catalytic pyrolysis of a PE plastic sample with and without activation revealed a notable difference. After undergoing thermal activation, a considerably larger number of pores were observed.

Regeneration of catalyst: As it was seen, after activation of char derived from catalytic pyrolysis of plastic wastes shows residue of MgO remaining in it which was confirmed in XRD analysis, which shows similar XRD diffraction pattern of the Ash-MgO catalyst showing a strong and sharp diffraction peak at 2θ angles of 28.3˚, 32.8˚, 39.5˚[26].

Conclusion

The liquid obtained from thermal and catalytic pyrolysis of PE was evaluated for its suitability as a fuel liquid using various fuel tests to determine its physical parameters. The physical parameters of the liquid, including density, specific gravity, viscosity, flash point and calorimetric values, were measured at different temperatures. It was observed that the liquid obtained from catalytic pyrolysis had physical parameters similar to diesel, with the exception of flash point. The gas chromatography-mass spectrometry (GCMS) analysis of the oil obtained from catalytic pyrolysis showed a hydrocarbon range of C_{10}-C_{15}, which is similar to the hydrocarbon range of diesel.

The char obtained after catalytic pyrolysis contained residual catalyst, which can be reused as a catalyst, thereby enabling the recovery of the catalyst. The use of a catalyst not only increased the yield of liquid but also reduced the reaction time of pyrolysis. Therefore, it can be concluded that catalytic pyrolysis is more advantageous than thermal pyrolysis.

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