Synthesis of Hydrocarbon Fuel by Thermal Catalytic Cracking of Polypropylene

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Abstract: The current study examined the thermal catalytic cracking reaction of polypropylene-based polymers to generate C_8-C_{18} hydrocarbons using metal oxide catalysts, including $MgCO_3$, $CaCO_3$, Al_2O_3, and MgO. A systematic analysis of the liquid productswas performed to determine the optimum catalyst that produces a highest yield of C_{10}-C_{12} hydrocarbon which falls under the gasoline range organics (GRO). Our data demonstrated that MgO produced the highest yield of liquid products compared to the other catalysts. The kinetic study suggeststhe potential mechanism of time and ratio (polymer vs. catalyst)-dependent polypropylene cracking. The hydrocarbon products were analyzed usingGC-MS and FT-IR to show that polypropylene cracking generates carbon constituents ranging from C_8 to C_{18} which is a range for naptha, petrol, kerosene, and diesel. The FT-IR spectra demonstrated that hydrocarbon products contain mostly alkene and alkane functional groups. A massof polypropylene waste plastic were converted into potential kerosene fuel using 10%metal oxide catalysts producing 85% liquid, 3% light gas and 12% residues with 25 minutesof reaction timeand a first order reaction kinetics.The calorimetric analysis showed that the liquid has a heating value within a range of 9.8–10.8Kcal/g.

Index Terms—catalyst, thermal catalytic cracking, polypropylene, metal oxide, GC-MS, FT-IR, waste polymers, kerosene fuel

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1 INTRODUCTION

An elevation in the amount of polymer wastes has been created due to the non-biodegradable nature and high increase in production of platics in the world. Most ofpolymer wastes are disposed in landfills or incinerated.Nevertheless, the polymer wastes are larger in size than the disparate organic residues and consequently occupy massive space in landfills and hence, the suitable way of getting rid of the plastics and incineration require high costs.In addition, incineration of waste plastics results in a negative environmental impact due to increased discharge of toxic compounds. For this reason, the need for a sustainable and renewable solutionof wasted polymers is required.

The carbon recycling by catalytic cracking is better alternative approach todictate the rise of polymer waste, since they are environmentally friendly when compared to incineration and disposal in landfills. Moreover, from carbon recycling, it is possible to re-

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gain energy and fuel while reducing the consumption of natural resources. The negative environmental impacts will be reduced when carbon products and energy are recovered.

Pyrolysis is a tertiary recycling process which could either be thermal or catalytic, and is developed in a desirable fashion to allow the conversion ofplastics into gas and liquid hydrocarbons. This is a procedure with proportionally low cost from which a wide distribution of products can be acquired. In pyrolysis, when heating occurs in the absence of oxygen, the organic compounds are decomposedto produce gaseous and liquid products, which can be used as fuels or sources of chemicals.

Pyrolysis can be enhanced by the annexation of catalysts, which will alter the rate of the chemical reaction by lowering the activation energy via reduction of the temperature and reaction time and allow the production of hydrocarbons with a higher added value. The use of catalysts gives an added quality to pyrolysis and cracking efficiency of catalysts depends on its chemical and physical properties. These specific characteristics enhance the rupturing of carbon bonds and determine the length of the chains of the products formed.

The cracking of polyprolylene produces propane gas which can be used as cooking gas. Polypropylene molecule is a straight and isotactic chain with the absence of crosslinking. Moreover, thermal degradation of polypropylene requires lower activation energy to break the C-H bond than polyethylene (PE) because carbon chain of polypropylene contains tertiary carbon atoms which have considerably lower resistance against degradation [13]. All of these factors make polypropylene to be selected as a starting material for the current study. Various catalysts have been used for plastics cracking including zeolites, alumina, etc. [14], [15], [16], [17]. However, there are many problems associated with these catalysts including high cost of production and operating conditions, including high temperature and more residence time for decreasing industrial feasibility [18], [19], [20], and [21].

Our study used MgO, $MgCO_3$, Al_2O_3, and $CaCO_3$ as catalysts to produce liquid hydrocarbon with low molecular weight hydrocarbons [22], [23], [24], [25], [25], and [26].Apart from production of liquid hydrocarbon, the current study revealed a potential mechanism and a fast catalyst recovery based on kinetic study. The current study examined the kinetics by collecting the liquid hydrocarbon after five minutes until the end of the reaction. Moreover, the temperature range for each reaction has been recorded in order to determine the optimum and maximum temperature for each catalyst. We aimed as follows: 1) To synthesize C_5-C_{22} hydrocarbons from polypropylene using several catalysts including MgO, $MgCO_3$, $CaCO_3$, and Al_2O_3, 2) To determine the kinetics and reaction mechanism, 3) To determine the reaction order and reaction rates, 4) To apply for the usage of the final products. Our data demostrated that the liquid products has a carbon range from C_8 to C_{18}which has proven that the liquid products can be used as kerosene for transportation, cooking, heating or jet fuel [27], [28], [29], [30].

2.1 Materials

Polypropylene was collected locally from Yola, Nigeria. Polypropylene was cleaned using liquid soap and water. Polypropylene was cut off into smaller pieces (1 x 1 cm)which wasplaced in round bottom flask(250ml). Selected metal oxide, including $CaCO_3$, $MgCO_3$, Al_2O_3, and MgO, were used as catalysts.

2.2 Experimental process converting polypropylene into low molecular liquid hydrocarbons

Polypropylene-based plastics (72 g) were cut and the shredded plastics were packed into a pre-weighed round bottom flask.Metal oxide catalyst (7.2 g, Al_2O_3, MgO, $MgCO_3$, and $CaCO_3$) wasalso packed inside the round bottom flask containing the feedstock. The masses of polypropylene and the catalysts were uniformly maintained through out the experiment. The glass reactor with known mass of feedstock (polypropylene + catalyst) was placed on a 200-watt heating mantle and heated to 350^oC. The reactor delivery tube was connected to a condenser leading to the receiving flask. The products were obtained as non-condensable gases, condensable liquid oil or wax, and solid residues. Condensable liquid oil and wax were collected into the beaker while the solid residues were leftover in the bottom of the reactor.

2.3 Analysis

The total percentage conversion was calculated as total % conversion= (initial weight of polypropylene - weight of unconverted polypropylene) x 100/initial weight of polypropylene. Thermo Nicolet IR100 FT-IR was used to determine the functional groups present in the liquid proudcts. The gas chromatography and mass spectroscopy (GC-MS) was used to determine the fragmentation pattern, potential functional group, and molecular weight.

2.4 Kinetic study

A fixed ratio of Polyporpylene (50 g) and each catalyst (5 g) was used for the kinetic study. To determine the liquid product yield over the time interval, liquid product was collected and measured each five minutes of the reaction until the end of the reaction. The reaction order was determined by plotting the graph of ln([liquid yield]/Δt) against time. Where [liquid] is liquid percentage yield expressed in percent and Δt is change of time which is 5minutes and time varied from 0 to 25 minutes. The reaction order, rate constant, reaction rate, and the activation energy were determined using four different catalysts.

3. Result and Discussion

We obtained the cracked products as in the form of uncondensated gases, liquid, and solid residues. The solid residuses were left in the flask, the liquids were collected in the preweighed beaker, and the gases were allowed to escape. Table1 illustrates theresults of liquid yield, mass of residues, gas evolved and total conversion yield obtained from the catalytic cracking of polypropylene (72 g).

Table 1

Percentage Yield of Products Using Different Catalyst Variation in Time

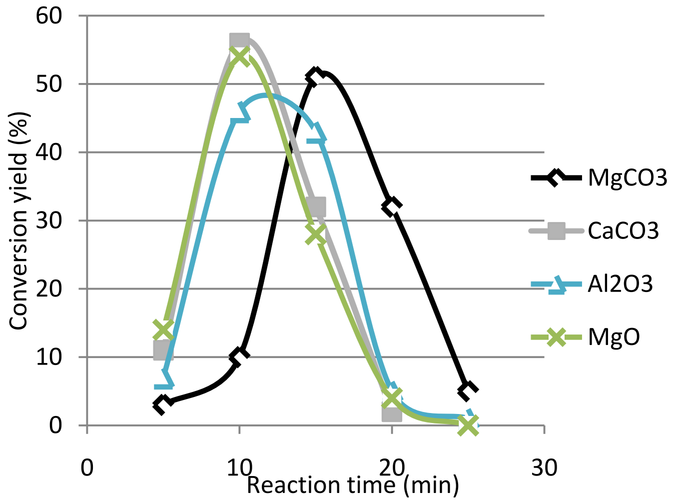
Mass of catalyst(g)	Time (min)	Liquid yield	Mass liquid yield (g)	Mass Residue (g)	Mass of gas evolved	Total Conversion (g), Yield
7.2g $MgCO_3$	0-5	1.14g	54g =73.5%	4g =5.2%	4.5g =20.3%	68.5g =94.4%
	5-10	4.24g				
	10-15	22.2g				
	15-20	14.0g				
	20-25	2.04g				
7.2g $CaCO_3$	0-5	4.50g	65.3g =90.5%	1.4g =1.9%	5.4g =7.5%	70.7g =98%
	5-10	24.2g				
	10-15	13.7g				
	15-20	0.89g				

	20-25	0.00g				
7.2g Al_2O_3	0-5	2.82g	64.9g =90.1%	2.8g =3.8%	4.8g =6.7%	69.7g =96.2%
	5-10	20.2g				
	10-15	18.6g				
	15-20	1.97g				
	20-25	0.48g				
7.2g MgO	0-5	6.39g	62.5g =86.3%	0.2g =0.3%	9.8g =13.5%	72.3g 99.7%
	5-10	24.19g				
	10-15	12.54g				
	15-20	1.77g				
	20-25	0.00g				

Our data demonstrated that $CaCO_3$ catalyst has the highest percentage yield (90.5%) followed by Al_2O_3, and MgO, while $MgCO_3$ has the lowest percentage yield (73.5%) for the condensed liquid (Table 1). $MgCO_3$ has the highest yield (5.2%) of solid products, followed by Al_2O_3 (3.8%), and $CaCO_3$(1.9%). MgO showed the lowest percentage yield (0.3%) of solid residues.$MgCO_3$ has the highest gas product yield while Al_2O_3 gives the lowest percentage yield of the gas products.

Figure 1

Conversion rate vs. reaction time



Based on the reaction rate, MgO demonstrated a highest catalyst efficiency to convert a large amount of feedstock polypropylene within a short period of time compared to the other catalysts that include $MgCO_3$, $CaCO_3$, and Al_2O_3. However, $MgCO_3$ showed a lowest catalytic ability for polypropylene cracking into liquid products compared to MgO, $CaCO_3$, and Al_2O_3.$MgCO_3$ was proved to be a functional catalyst for higher gas production (20.3%) during degradation. Our data demonstrated that the catalytic efficiencies were in the order of $MgO>CaCO_3>Al_2O_3>MgCO_3$.

The liquid yield in every 5 minutes of the reaction using the various catalysts showed the potential mechanism as time and ratio-dependent first order reaction in polypropylene cracking (Figure 1). $CaCO_3$ has thehighest liquid conversion rate between 10–15 minutes of reaction time. The hydrocarbon liquid products were characterized using FT-IR for functional group analysis.

Table 2

FT-IR spectra showing functional groups in polypropylene cracking products

Wavenumber (cm^{-1})	Bond, Functional group
2950-2870	alkane
1650-890	alkene
740	alkene, phenyl ring substitution

2110 – 1160	alcohol, ethers, carboxylic acids, or esters
16940,1710,1770	aldehydes, ketones, carboxylic acids, or esters

IR data indicates the presence of different hydrocarbons as alkanes and alkenes. In addition, part of oxygenated compounds, inlcuding alcohol, ketones, and esters are also found in the liquid products, which may be due to oxidative degradation of polypropylene in the presence of oxygen in the reactor.

Table 3

Carbon range as a function of reaction time

Carbon range	Reaction time(min.)
Gasoline(C_5-C_{12})	0 – 11
Diesel(C_{12} – C_{20})	11 -23
Viscous residues (> of C_{20})	>23

FT-IR analysis demonstrated that the liquid products from all of the four catalysts had the following chemical bonds, inlcuding cis-RCH=CHR, =C-H (trans), $=CH_2$ (trans), $-CH_3$ bending, -C=C-, C_{sp3}-H and C_{sp2}-H.Gas chromatography and mass spectrometry (GC-MS) showed the carbon numbers present in the liquid products from each catalyst. It was concluded that the liquids products obtained from polypropylene cracking using $MgCO_3$, $CaCO_3$, MgO, and Al_2O_3 catalysts were carbon ranged from C_8 to C_{18}.

Thermal catalytic cracking and the reaction mechanism

A potential mechanism was derivedbased on GC-MS results for the thermal cracking of polyproptlene, consisting three possible processes that include C—C bond breaking, C—H bond breaking, then hydrogenation. C-C bond cleavage may occur either on each carbon-carbon bond (C—C, 82.94 Kcal/mol) or on carbon-hydrogen (CH_3—H, 101.10 Kcal/mol, CH_2—H, 439.00 Kcal/mol, CH—H, 107.01 Kcal/mol and C—H, 83.00 Kcal/mol. The average C—H bonding energy is 98.09 Kcal/mol. The cleavage of these bonds may resultin a variety of products as shown in our Table 4. Due to the higher bonding energy of the C—H bond, the possibility of breaking C—H bond is lower than that of the C—C bond breakage. High temperatures lead to the formation of free radicals, which react with hydrocarbons, producing new hydrocarbons and new free radicals as shown in the following mechanism.

$$R—CH_2—CH_2—CH_2—CH_3+\bullet CH_3 \rightarrow R—CH_2—CH_2—CH_2—CH_2\bullet+CH_4$$

Then free radicals could be decomposed to generate olefins and new radicals.

$$CH_2—CH_2—CH_2—CH\bullet—CH_2—CH_2— \rightarrow R—CH{=}CH_2+CH_3—CH_2\bullet$$

Polypropylene cracking takes place through the end-chain scission or depolymerization, producing monomers, dimers or oligomers with random chain scission breaking down into smaller fragments of various lengths of C—C bond. The C—C bond cleavage may occur at any position, however, CH_3—CH_2-bond breaking shows higher possibilities than that of —CH_2—CH_2— breaking. The least possibility is the breaking of C—H bond. Hydrogen generated from this mechanism may react with alkenes to generate alkanes. Unsaturated longer chain hydrocarbons could be more easily hydrogenated than shorter chain ones because longer chain hydrocarbons are mostly saturated alkanes.Our data reveals that thermal cracking of polypropylene may occur within 150-300°C temperature ranges.A kinetic model of polypropylene cracking reaction was determined and the rate constants for the reactions were studied under the assumption that no mass transfer resistance happens inside and outside of catalyst powder and no catalyst deactivation occurs inside the rotating basket. The kinetic model was used as there are hundred of components involved in the products. Experimental result shows that every reaction is the first order with respect to the reactant. The polypropylene degradation with 10% of catalyst diminishes the activation energy in an isoconversion of 95%, confirming the influence of the catalyst on the activation energy. We noted that the degradation time diminishes significantly when the reaction temperature was increased. The activation energy of the catalytic cracking reaction was calculated in the range of 13.2 Kcal/mol and24.3 Kcal/mol.

Table 4

GC-MS analysis showing product name, formula, molecular weight and % composition

Composition produced by Al_2O_3				**Composition produced by $MgCO_3$**			
Compound Name	Formula	Molecular Weight	Percentage Composition	Compound Name	Formula	Molecular Weight	Percentage Composition
2,4-hexadiene	C_6H_{10}	82	1.33	1,-heptene,5-methyl	C_8H_{16}	112	7.76
1,4-hexadiene, 5-methyl	C_7H_{12}	96	0.23	Cyclohexane, 1,2,4-trimethyl	C_9H_{12}	120	1.23
Heptane, 4-methyl	C_8H_{18}	114	0.96	2,3,3-trimethyl-1-hexene	C_9H_{19}	127	1.08
1,1,3,4-tetramethylcyclohexane	$C_{10}H_{20}$	140	0.31	Nonane, 4-methyl-	$C_{10}H_{22}$	142	0.74
1-heptene, 5-methyl	C_8H_{16}	112	4.81	1-hexene, 3,3-dimethyl	C_8H_{16}	112	1.65
2,4,6-trimethyl-3-heptene	$C_{10}H_{21}$	141	1.54	1-methylpentyl cyclopropane	$C_{11}H_{17}$	149	0.69
Nonane, 4-methyl	$C_{10}H_{21}$	141	1.54	4-decene, 3-methy-	$C_{11}H_{22}$	154	0.71
1-hexene, 3,3-dimethyl	C_8H_{16}	112	1.00	1-decene,8-methyl	$C_{11}H_{22}$	154	8.65
Decane, 4-methyl	$C_{11}H_{24}$	156	1.56	2.2-dimethyl-3-heptene trans	C_9H_{18}	126	1.95
2,4,6-trimethyl-3-heptene	$C_{10}H_{21}$	141	2.83	1,7-nonadiene, 4,8-dimethyl	$C_{11}H_{20}$	152	2.84
1-decene, 8-methyl	$C_{11}H_{21}$	153	2.53	Cyclohexane, 2,4-diethyl-1-methyl	$C_{11}H_{22}$	154	1.26
Cyclohexane, 1,2-diethyl-3-methyl	$C_{11}H_{22}$	154	3.23	2-undecene, 2-methyl	$C_{12}H_{24}$	168	1.10
3-hexene,2,2,5,5-tetramethyl	$C_{10}H_{20}$	140	3.03	Octadecane	$C_{18}H_{40}$	256	1.85
Cyclohexane, 1,1,2-trimethyl	C_9H_{18}	126	6.34	Octadecene	$C_{18}H_{36}$	252	4.02
1,5-Octadiene, 7-methyl	C_9H_{15}	123	2.66	1-pentene, 2,3-dimethyl	C_7H_{12}	96	22.3

Cyclopentane,(2-methylpropyl)	C_6H_{10}	82	5.24	3-decene, 2,2-dimethyl-, (E)	$C_{12}H_{24}$	168	1.75
3-decene,2,2-dimethyl-, (E)	$C_{14}H_{24}$	192	5.12	Pentadecane	$C_{15}H_{32}$	212	5.45
Cyclohexane,eicosyl	C_6H_{12}	84	3.40	1,6-Octanadiene, 2,5-dimethyl- (E)	$C_{10}H_{19}$	139	5.28
3-decene,2,2-dimethyl- (E)	$C_{14}H_{24}$	192	19.6	Cyclopentane,(2-methylpropyl)	C_5H_{10}	70	16.25
Cyclopentane,1-methyl-3	C_9H_{18}	126	21.23				

Composition produced by MgO				**Composition produced by $CaCO_3$**			
Compound Name	Formula	Molecular Weight	Percentage Composition	Compound Name	Formula	Molecular Weight	Percentage Composition
2,4-dimethyl1,4-pentadiene	C_7H_{12}	96	10.56	1,4-hexadiene,5-methyl	C_7H_{12}	96	0.23
1,4-Hexadiene, 5-methyl-	C_7H_{12}	96	0.53	Heptane, 4-methyl	C_8H_{18}	114	1.57
4-methyl-2-heptene	C_8H_{16}	112	1.08	1-heptene, 2-methyl-	C_8H_{17}	113	1.04
Heptane, 4-methyl-	C_8H_{18}	114	2.18	2,4,6-trimethyl-3-heptene	$C_{10}H_{20}$	130	0.72
1-Heptene, 2-methyl-	C_8H_{16}	112	2.22	1-heptene, 5-methyl-	C_8H_{16}	112	13.90
1-hexene,3,3,5-trimethyl-	C_9H_{18}	126	1.31	Cyclohexane,1,2,4-trimethyl	C_9H_{18}	126	2.46
1-heptene, 5-methyl-	C_8H_{16}	112	24.60	Tetramethylcyclopentane	C_8H_{16}	124	1.70
Cyclohexane, 1,2,4-trimethyl-	C_9H_{18}	126	4.43	1,6-octadiene, 2,5-dimethyl-(E)	$C_{10}H_{18}$	138	0.87
Tetramethylcyclopentane	C_9H_{18}	126	2.21	Nonane,4-methyl-	$C_{10}H_{22}$	132	0.91
1,6-heptadiene, 2,3,6-trimethyl-	$C_{10}H_{18}$	138	1.18	2-decene, 4-methyl- (Z)	$C_{11}H_{21}$	153	1.16
Nonane, 4-methy-	$C_{10}H_{21}$	141	1.01	Decane, 4-methyl-	$C_{11}H_{24}$	156	1.66
2-decene, 7-methyl- (Z)	$C_{11}H_{22}$	154	1.48	2-decene, 5-methyl- (Z)	$C_{11}H_{22}$	154	0.42
Decane, 4-methyl-	$C_{11}H_{24}$	156	2.01	2-decene, 9-methyl- (Z)-	$C_{11}H_{22}$	154	0.50
4-decene, 2-methyl-	$C_{11}H_{22}$	154	0.78	3-decene, 2,2-dimethyl-, (E)	$C_{12}H_{24}$	156	0.67
Cyclopentane, (1,3-dimet hyl-butyl)-	C_5H_{10}	70	0.69	1-decene, 8-methyl-	$C_{11}H_{22}$	154	6.78
1-undecene, 8-methyl	$C_{12}H_{24}$	168	9.54	Cyclopentane, propy	C_8H_{16}	112	1.49
2,2-dimethy-3-heptene trans	C_7H_{14}	98	1.69	1,7-nonadiene, 4,8-dimethyl-	$C_{11}H_{20}$	152	2.14
1,7-nonadiene, 4,8-dimethyl	$C_{11}H_{20}$	152	2.84	Cyclohexane, 2,4-diethyl-1-methyl	$C_{11}H_{21}$	153	1.21
Cyclopentane, 1,2-diethyl-3-methyl-	$C_{10}H_{20}$	140	1.22	2-undecene, 2,-methyl-	$C_{12}H_{24}$	168	1.01
4-nonene, 2,3,3-trimethyl-, (Z)	$C_{12}H_{24}$	168	1.08	Cyclopentane, (2-methylbutyl)-	C_5H_{10}	70	1.18
Cyclopentane, (2-methylbutyl)-	C_5H_{10}	70	1.17	2-pentene, 3-methyl- (Z)-	C_6H_{12}	84	7.31
5-undecene, (E)	$C_{11}H_{22}$	154	0.55	3-hexene, 2,2,5,5-tetramethyl, (Z)	$C_{10}H_{20}$	140	1.46
5-octadecene,-(E)	$C_{18}H_{36}$	252	2.14	Hexadecane	$C_{16}H_{34}$	$C_{16}H_{34}$	3.85
Cyclohexane, 2,4-diethyl-1-methyl-	$C_{11}H_{22}$	154	11.64	Dodecane, 2,4,6-trimethyl	$C_{15}H_{32}$	212	6.46
1-pentene, 2,3-dimethyl-	C_7H_{14}	98	10.49	1-butene, 3,3-dimethyl-	C_6H_{12}	84	16.61

Our cracking reaction required a high amount of activation energy to break down the polypropylene molecules when $CaCO_3$ was used as catalyst. However, Al_2O_3 was able to break polypropylene at lowest activation energy compared to other catalysts. The GC-MS showed that the liquid yield has the carbon range from C_8 to C_{18} which is a range for naptha, petrol, kerosene (paraffin) and diesel. The various fractions could be used as fuel for car engines, solvents in petrol, fuel for aircraft and stoves, fuel for road vehicles and trains. The liquid products were a mixture of aliphatic and cyclic liquid hydrocarbons. Based on FT-IR, the majority of molecules present in the liquid products were unsaturated alkenes.This is partly because polypropylene cracking reactions include C—H bond breakage, introducing unsaturation, especially using metal oxide catalysts. The calorimetric analysis showed that the liquid has a heating value within a range of 9.8– 10.8Kcal/g. This energy could be increased further at an optimum set temperature and heating rate.

Conclusion

The current study introduced different catalysts in converting the waste plastics into desired fuels. It was demonstrated that $CaCO_3$ catalyst has the highest liquid percentage yield of 90%,while $MgCO_3$catalyst has the highest yield of 5.2% of solid and gas product yield. Moreover, MgO catalyst has the highest degradation conversion rate; Al_2O_3 initiated cracking reaction with lowest activation energy but with lower yield. The kinetic study demonstrated that all cracking reactions are first order with respect to the reactant as a result of the isoconversion method. The cracked liquid products might be used in industrial boiler, burners, power generators, automobiles and jet fuel. This process will bring possible solutions in protecting and preserving our environment and the ecosystem. Furthermore, our technology will create new opportunities forthe thermal catalytic process to reduce polymer waste from the environment and enhancecarbon/energy recycling strategy.

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