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Catalytic cracking of polyethylene plastic waste using synthesised zeolite Y from Nigerian kaolin deposit

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Abstract

The increasing rate of accumulation of plastic waste (PW) is quite disturbing to the world, particularly in developing nations due to its non-biodegradable nature and inadequate waste management practices. The need to properly manage this waste and utilize the potential and chemical energy value that can be derived from this waste justifies the encouragement and employment of newer and better recycling methods and technology of these wastes. Therefore, this has led us to explore the catalytic pyrolysis of plastic waste using zeolite Y synthesized from kaolin deposit in Covenant University, Sango Ota, Ogun state of Nigeria. A stainless steel packed bed reactor was used in the cracking of low-density polyethylene (LDPE) plastic wastes into liquid fuel components at a temperature of 300 °C using zeolite Y catalyst. The liquid fuel obtained from the catalytic pyrolysis was analyzed using GC–MS. Fifty compounds were identified, which revealed the presence of mostly alkenes and aromatics in the hydrocarbons range of C_8 – C_{29} . This is made up of 56% of gasoline fractions range of C_6 – C_{12} , 26% of diesel and kerosene fractions range C_{13} – C_{18} , and 10% of fuel oil range C_{18} – C_{23} , while 8% is residual fuel range greater than C_{24} .

Keywords Polyethylene (PE) \cdot Catalytic cracking \cdot Pyrolysis \cdot Zeolite Y \cdot GC–MS

Introduction

The use of plastic material is of greater importance in our daily lives and its consumption has led to the drastic increase of PW in the second half of the twentieth century [1]. The demand for plastics has increased over time because they are considered as alternative materials providing solutions to various sectors such as aerospace, engineering, medicals, and electronics [2]. Polyethylene plastics are nonbiodegradable, lightweight, and flexible materials and are extremely important for technology advancement due to these properties.

PW is the major constituent of municipal solid waste (MSW) and seen as fast approaching one of the largest MSWs in the developing countries [3]. The accumulation

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¹ Department of Chemical Engineering, College of Engineering, Covenant University, Ota, Nigeria of this PW over a length of time in conjunction with the improper and conventional waste management strategies have led to major health and environmental hazards such as greenhouse gas emissions, groundwater pollution, and several other human health problems [4]. In addition, developing countries do not consider the advantages of economic gain involved by utilizing certain recycling methods, but still depend solely on the conventional method of landfilling of MSW disposal.

Hence, effort toward the conversion of this PW to resourceful materials of valuable products and energy, in addition to solving the shortage of natural resources, in the nearest future has been a major area of research [2]. This can be achieved by exploring recycling methods, which so far is the best option for PW management [5]. PW recycling methods have been mainly grouped into four major types, namely primary recycling involves waste scraps being processed into products with similar properties to the original products; secondary recycling has to do with waste/scrap plastics being processed into materials that have different properties to that of the initial product; tertiary methods deal with use of these waste scraps in the production of essential fuels and chemicals or as a segregated waste; and lastly quaternary recycling involves the burning of these plastics



and retrieving energy contents afterward [6]. Each of these recycling methods is advantageous and effective for different applications.

Thermoplastics and thermosets are the main types of plastic used for daily consumptions. Upon the introduction of heat, thermoplastics become soft and later harden up, while thermosets harden irreversibly. There are six main components of MSW, namely: polyethylene (PE), there are two major types and make up about 40% of the MSW which are low-density polyethylene (LDPE) and high-density polyethylene (HDPE), polyethylene terephthalate (PET), polyvinyl chloride (PVC), polystyrene (PS), and polypropylene (PP) [2, 7].

Thermal cracking, or pyrolysis, involves the degradation of the long chain polymeric materials by heating in the absence of oxygen. Gas, oil, and char are the three major products obtained during the pyrolysis process and these products are of great value to the production and refinery industries [8]. Compared to other MSW management practices, pyrolysis is more desirable and satisfactory environmentally because it reduces the carbon footprint of products. Pyrolysis process also lessens the amount of carbon monoxide and carbon dioxide emissions by making effective use of an inert atmosphere that is free of oxygen to avoid the formation of dioxins through product reaction with oxygen [9, 10]. This pyrolysis process is a cleaner technology, a drive toward energy security and a measure to combat fossil fuel depletion.

Zeolites are crystalline microporous aluminosilicate materials with an extra framework having ion exchange capabilities and thermal stability over a wide temperature range. They are currently used as catalysts for catalytic cracking of heavy hydrocarbons in many commercial processes; zeolite Y is a highly porous faujasite zeolite with Si/Al ratio of 1.5–3.8 [11, 12].

Zeolites are produced both naturally and synthetically; however, those that find applications in the petroleum and petrochemical industries are mainly the synthetic ones. The synthesis of zeolites from clay as a source of silica and alumina is widely known and has yielded great achievement [13, 14]. Arobieye clay in Nigeria was found to have a cost advantage of 25% over the commercial chemical in the synthesis of zeolite. This kaolin clay is in abundance in many parts of Nigeria and previous works have shown that it is a rich alternative source for obtaining alumina [15].

There have been a number of published articles on the catalytic degradation of PE plastics, particularly the LDPE. For example, Neves et al. [1] undertook the catalytic pyrolysis of polyethylene using zeolite. Almeida et al. [16] also investigated the thermal and catalytic pyrolysis of PW and obtained products with a high added value such as fuel oils

and feedstock for new products. Kunwar et al. [17] investigated the catalytic and thermal depolymerization of lowvalue post-consumer high-density polyethylene plastic. Thermal cracking yielded mainly diesel and vacuum gas oil fractions, while catalytic cracking (using Y zeolite and MgCO₃) yielded increased gasoline and diesel-range fractions. The fuel properties of the diesel-range distillate were found comparable to those of ultralow sulfur diesel (ULSD). Adeoye et al. [18] synthesized zeolite Y with Si/Al ratio of 3.22 from Arobieye clay in Nigeria and further tested its catalytic effect on cyclohexane.

This work seeks to further investigate and evaluate the activity of this zeolite Y on the cracking of LDPE PW. The success of this work will enhance the effectiveness of plastic waste processing to useful products in Nigeria and Africa in general in an economically affordable manner and hence reduce the environmental hazard posed by their indiscriminate dumping.

Experimental

Materials

LDPE was used in this experiment. The PPW was obtained from waste bins and garbage from Ota, south-west Nigeria. It was then sorted, cleaned to remove dirt, and then shredded into smaller particle sizes. The catalyst used was a Covenant University pilot plant-synthesized zeolite Y from Arobieye kaolin deposit in Nigeria with an Si/Al ratio of 1.45 which is rich in oxides of calcium, iron, and titanium.

Characterization of catalyst used

X-ray diffraction (XRD) patterns were recorded on an X-ray diffraction machines to know the relative crystallinity and crystal structure of synthesized zeolite using a Cu–Ka radiation with the wavelength of 1.540598. Scanning electron micrograph (SEM) was analyzed by phenom prox SEM with the model number 800-07334 and part number MVE0224651193 used to determine the morphology of the zeolite.

The chemical composition of both the raw Arobieye kaolin and synthesized zeolite was analyzed using X-ray fluorescence (XRF).

Experimental procedures

LDPE was collected and prepared by cutting into smaller particle sizes of 3–4 mm and charged into a packed bed pyrolysis reactor which was operated at a temperature of



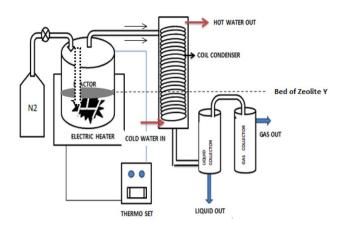


Fig. 1 Schematic representation of the pyrolysis setup. Source: Ajibola et al. (this study)

 $300 \,^{\circ}$ C and at atmospheric pressure. As shown in Fig. 1, the reactor is a lagged cylindrically shaped stainless steel vessel of capacity 300 ml with a height of 11 cm and diameter of 6 cm. The reactor was electrically heated with a 2 kW heating element equipped with an automatic temperature controller and connected to a coil condenser, and subsequently liquid and gas collectors.

100 g of polymer to 10 g of catalyst was used in the experiment and fed into the reactor, tightly closed, and purged with nitrogen gas at the start of each experiment at the rate of 20 ml/min for 10 min. The heating element was turned on and regulated to a specified temperature to liquefy the PPW. The liquefied PPW at the set temperature was vaporized. The vaporized PPW passed through the catalyst bed where reaction occurred. The vaporized product was finally condensed and collected for analysis. The solid (S), gaseous (G), and liquid product (L) yields were calculated using the formula given below as shown in Eqs. 1, 2, 3 (Source: Patil [19]):

$$L (wt\%) = \frac{\text{weight of liquid product}}{\text{weight of plastic feed}} \times 100,$$
(1)

$$S (wt\%) = \frac{\text{weight of solid product}}{\text{weight of plastic feed}} \times 100,$$
 (2)

$$G (wt\%) = 100 - (L (wt\%) + S (wt\%)).$$
 (3)

Analytical methods

The pyrolyzed oil was analyzed using gas chromatography-mass spectrometry (GC-MS) to determine the chemical constituents of the hydrocarbon products. The GC-MS and column oven temperature conditions used are stated in Table 1.

Identification of components

Mass spectrum GC–MS was interpreted using the National Institute Standard and Technology (NIST) library of mass spectra database. Comparison of the unknown and known spectrum components was done using the NIST library. The molecular name and weight of the compounds were also confirmed.

Results and discussion

Table 2 shows the XRF analysis of the synthesized zeolite Y, which shows SiO_2/Al_2O_3 of 3.22 in comparison with the chemical composition of the raw Arobieye kaolin used which shows a SiO_2/Al_2O_3 of 1.45. These results show that the Si/Al ratios are within the expected ranges of 1–2 and 1.5–3.8, respectively [12, 20]. Figure 2 shows the XRD pattern of the synthesized zeolite Y with peaks observed at Bragg's angles of 14, 19, 21, 24, 26.8, 31.9, 34.9, 39.9, 49.8, and 51° [21]. Figure 3 also shows the structural morphology of the synthesized zeolite at a magnification of 3500 depicting clear crystal formation of zeolite Y from the structural boundaries and shapes.

The results from the GC–MS analysis established the identity of the number of compounds or fractions of the liquid fuel obtained from the cracked PPW. Identification of compounds was also done through mass spectrometry attached with GC. Figure 4 and Table 3 show the yield of the pyrolyzed PPW at a temperature of 300 °C. From this result, it can be seen that the liquid yield is more than that of other products. The liquid fuel extracts were yellow in color.

Characterization of liquid products

The pyrolyzed PPW reveals the presence of hydrocarbons in the C₈–C₂₉ range. Analyzing the compounds present in the liquid fuel of the catalytically pyrolyzed LDPE, it can be deduced from Fig. 5 that the carbon number distribution of LDPE contains the gasoline (C₆–C₁₂) fraction of 56%, diesel and kerosene (C₁₃–C₁₈) fraction of 26%, and fuel oil (C₁₉–C₂₃₎ fraction of 10%, while the residual fuel oil (>C₂₄) fraction range was 6%.

Although the functional group in this present study was not detected. From literature review, the most abundant



 Table 1 GC/MS condition. Source: Ajibola et al. (this study)

GC–MS Agilent 433UI HP-5 ms ultra inert

Column oven temperature	50 °C
Injection mode	Split
Injection temperature	150 °C
Injection volume	1 ml
Split ratio	10:1
Average velocity	36.445 cm/sec
Column pressure	7.6522 psi
Column flow	1.2211 ml/min
Carrier gas	Helium 99.9995% purit
Frequency	50 Hz
Electron multiplier volts	1024.9

Rate (°C/min)	Temperature (°C)		Hold time (min)
_	50		0
8	300		9
Column			
Length		30.0 m	
Diameter		250 µm	
Film thickness		0.25 µm	
Total runtime		32 min	
MS conditions			
Source temperature			230 °C
Start mass range m/z			50
End mass range m/z			550

Table 2	XRF	analysis	of	the	raw	Arobieye	kaolin	and	synthesized
zeolite.	Sourc	e: Adeoye	e et	al. [18]				

Chemical composition	Raw Arobieye kaolin	Synthesized zeolite Y
SiO ₂	48.79	39.71
Al_2O_3	33.58	20.94
Fe ₂ O ₃	2.65	2.26
CaO	2.98	3.15
MgO	0.58	0.28
SO ₃	0.04	6.76
Na ₂ O	0.07	28.47
K ₂ O	0.01	0.06
TiO ₂	1.48	1.67
P_2O_5	0.10	0.07
Mn ₂ O ₃	0.01	0.02
Si/Al (wt%)	1.45	3.22

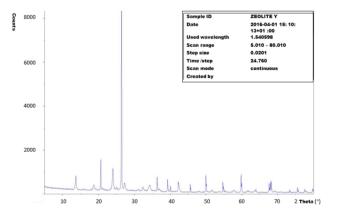


Fig. 2 XRD pattern for the synthesized zeolite Y. Source: Adeoye et al. [18]



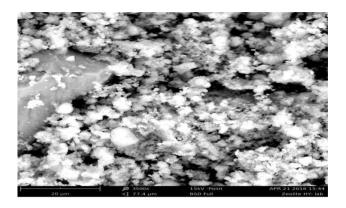


Fig.3 SEM images for the synthesized zeolite Y. Source: Adeoye et al. [18]

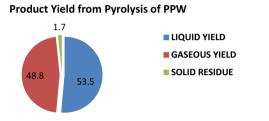


Fig.4 Product yield of pyrolyzed PPW at temperature of 300 °C. Source: Ajibola et al. (this study)

Table 3 Yields of liquid, gas, and solid residue from pyrolyzed PPW.Source: Ajibola et al. (this study)

Products	Yield wt%	Hydrocarbon ranges	Yield wt%
Liquid	53.5	C ₆ -C ₁₂	56
		$C_{13} - C_{18}$	26
		C ₁₉ -C ₂₄	10
		> C ₁₂	6
Gaseous	44.8		
Solid residue	1.7		

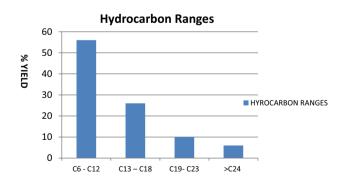


Fig.5 Hydrocarbon ranges of the liquid fuel composition. Source: Ajibola et al. (this study)

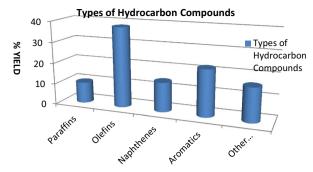


Fig. 6 Functional groups of the liquid fuel composition. Source: Ajibola et al. (this study)

groups were alkenes, alkanes, and aromatics [19]. From Fig. 6, the compound obtained from the pyrolyzed PPW shows the presence of several functional groups such as paraffins, olefins, naphthenes, aromatics, aldehydes, and esters. It shows that olefins and aromatics are the major compounds present [22, 23].

This present study reveals that the catalytic pyrolysis of LDPE plastic has yielded a higher fraction of gaseous product and lesser liquid fraction when compared to our previous work on non-catalytic thermal cracking of this plastic waste. This also agrees with the work of Seo et al. [24] that showed an increase of gaseous product from 13 to 63.5% using a catalyst for the pyrolysis reaction.

Fifty compounds were detected as shown in Table 4, and the five major compounds confirmed were styrene (4.96%), cyclopropane, nonyl (4.65%), indecene (4.32%), Z-5-nonadecene (3.73%), and indene (3.02%) with the retention times of 4.391, 7.27, 11.269, 20.104, and 6.909 min, respectively. The compounds obtained here were similar to those obtained by Patil et al. [19].

Conclusion

The catalytic cracking of the LDPE plastic wastes using zeolite Y has revealed the great usefulness of this waste as a rich source of raw materials for the production of a variety of chemicals and a good alternative to the production of automobile fuel.

GC–MS showed the compositional analysis of the liquid fuel obtained containing mainly aliphatic and aromatic compounds in the carbon range of C_8 – C_{29} . The liquid products obtained have similar products to fossil fuels and can, therefore, be used as alternative fuels for a more sustainable



 Table 4
 Chemical compositions of the liquid fuel products obtained from the catalytic pyrolysis of LDPE

Peak	eak Retention time Area %		Compound	Molecular formula	Molecular weight	
	3.625 1.21		2,4-dimethyl-1-heptene	C ₉ H ₁₈	126.2392	
	3.916	1.36	Benzene, 1,3-dimethyl-	$C_{8}H_{10}$	106.1650	
	4.037	1.25	Benzene, 1,3-dimethyl-	$C_{8}H_{10}$	106.1650	
	4.391	4.96	Styrene	C ₈ H ₈	104.1491	
	4.62	1.08	1-(2-propenyl)cyclopentene	$C_{8}H_{12}$	108.1809	
	5.187	1.35	1-(2-propenyl)cyclopentene	$C_{8}H_{12}$	108.1809	
	5.45	1.06	Benzene, 1-ethyl-4-methyl-	C_9H_{12}	120.1916	
	5.765	2.01	1,9-Decadiene	$C_{10}H_{18}$	138.2499	
	5.982	2.92	1-Decene	$C_{10}H_{20}$	140.2658	
0	6.051	1	Bicyclo[4.2.0]octa-1,3,5-triene- methyl	C ₈ H ₈	104.1491	
1	6.554	1.04	Bicyclo[4.2.0]octa-1,3,5-triene	C ₈ H ₈	104.1491	
2	6.749	1.13	Indane	C_9H_{10}	118.1757	
3	6.909	3.02	Indene	C ₉ H ₈	116.1598	
4	7.447	0.99	Bicyclo[4.1.0]heptane,3-methyl	$C_{8}H_{14}$	110.1095	
5	7.55	2.73	5-Undecyne	$C_{11}H_{20}$	152.2765	
6	7.27	4.65	Cyclopropane, nonyl-	$C_{12}H_{24}$	168.3190	
7	7.859	1.22	Undecane	$C_{11}H_{24}$	156.3083	
8	8.809	2.87	2-Methylindene	$C_{10}H_{10}$	130.1864	
9	8.906	1.87	2-Methylindene	$C_{10}H_{10}$	130.1864	
0	9.072	0.99	Tetracyclo[5.3.0.0<2,6>.0<3,10>]deca-4,8-di- ene	$C_{10}H_{10}$	130.1864	
1	9.341	1.8	1,11-Dodecadiene	C ₁₂ H ₂₂	166.3031	
2	9.461	2.46	Azulene	$C_{10}H_{8}$	128.1705	
3	9.667	1.12	Dodecane	$C_{12}H_{26}$	170.3348	
4	9.724	1.07	2-Dodecene, (Z)-	$C_{12}H_{24}$	168.3190	
5	10.846	1.11	1H-indene, 1,3-dimethyl-	C ₁₁ H ₁₂	144.2130	
6	11.092	2.62	17-Octadecynoic acid	$C_{18}H_{32}O_2$	280.4455	
7	11.269	4.32	1-Tridecene	C ₁₃ H ₂₆	182.3455	
8	11.389	2.21	Naphthalene, 1-methyl-	$C_{11}H_{10}$	142.1971	
9	11.693	1.64	Naphthalene, 2-methyl-	$C_{11}H_{10}$	142.1971	
0	11.778	1.22	Cyclohexasiloxane, dodecamethyl-	$C_{12}H_{36}O_6Si_6$	444.9236	
1	12.763	2.03	1,13-Tetradecadiene	$C_{14}H_{26}$	194.3562	
2	12.946	2.95	1-Tetradecene	$C_{14}H_{28}$	196.3721	
3	13.804	1.21	Naphthalene, 1,6-dimethyl-	$C_{12}H_{12}$	156.2237	
4	14.37	1.7	cis-9-Tetradecen-1-ol	$C_{14}H_{28}O$	212.3715	
5	14.502	2.21	Chloroacetic acid, pentadecyl este	C ₁₇ H ₃₃ ClO ₂	304.899	
6	15.893	1.9	Pentadecanal-	C ₁₅ H ₃₀ O	226.3981	
7	17.323	1.77	cis-9-Tetradecen-1-ol	$C_{14}H_{28}O$	212.3715	
8	17.455	1.54	3-Heptadecene, (Z)-	$C_{17}H_{34}$	238.452	
9	18.799	1.26	1-Octadecene	$C_{18}H_{36}$	252.4874	
0	19.984	1.11	Oxirane, hexadecyl-	C ₁₈ H ₃₆ O	268.4778	
1	20.104	3.44	Z-5-Nonadecene	C ₁₉ H ₃₈	266.513	
2	22.507	2.9	1-Nonadecene	$C_{19}H_{38}$	266.513	
3	23.657	2.25	1-Nonadecene	C ₁₉ H ₃₈	266.513	
4	24.704	2.04	9-Tricosene, (Z)-	$C_{23}H_{46}$	322.621	
5	25.746	2.5	Cyclotetracosane	$C_{24}H_{48}$	336.6379	
6	26.724	1.51	1-Nonadecene	C ₁₉ H ₃₈	266.513	
7	28.59	2.95	Octadecane	C ₁₈ H ₃₈	254.4943	
8	29.448	2.46	Tetracosane	C ₂₄ H ₅₀	338.6538	
9	30.278	1.85	Nonacosane	C29H60	408.7867	
50	31.09	1.33	Hexadecane, 1-iodo-	C ₁₆ H ₃₃ I	352.3377	



and cleaner environment when necessary blending is done for upgrading.

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