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Recycling Post- Consumer Waste from Low Density Polyethylene Via Low Temperature Catalytic Pyrolysis

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ABSTRACT: Pyrolysis with or without the aid of catalyst have attracted much attention for the conversion of waste plastics to their monomers or partially de-polymerized oligomers. The resulting monomers can be recycled to the original or related polymeric product as well as gasoline range products. In this work, the pyrolysis of contaminated Low Density Polyethylene (LDPE) was done both thermally and catalytically. The gases from each sample were collected in Tedlar bags and analysis was done using a Buck 530 Gas chromatograph. Results from the thermal pyrolysis showed aliphatic hydrocarbons within the range of $C_1 – C_9$ with a total concentration of 36.0906 ppm and 64.5041 ppm for gases collected at 200 ºC and 350 ºC respectively. The pyrolysis was also repeated under the influence of zeolite catalyst using catalyst/sample ratios of 1:8 and 1:16 at 150 ºC and 250 ºC. Results revealed total yield of gases for LDPE under the zeolitic effect of temperatures of 150 ºC and 250 ºC using catalyst/sample ratio of 1:8 to be 721.0371 ppm and 835.0906 ppm. The corresponding values obtained at 150 ºC and 250 ºC using catalyst/sample ratio of 1:16 were 697.4464 ppm and 713.7277 ppm respectively. The hydrocarbon gases revealed mainly $C_1– C_{10}$ aliphatic hydrocarbons. These when fractionated can result into combustible gases and gasoline range product.

KEY WORDS: combustible gases, Pyrolysis, low density polyethylene, zeolite.

INTRODUCTION

Plastics are materials that offer fundamental contribution to the society due to their versatility and relatively low cost. As a result of this, large amount of plastic waste is generated due to increase in its usage each year. This increase in the amount of plastic waste does cause some environmental problems since plastics do not degrade quickly and can remain in the environment for a long time. A larger part of this waste is disposed of in land fills or is incinerated [1-4].

Their destruction by incineration or use in land filling pose serious air pollution problems due to the release of airborne particulates and carbon (iv) oxide into the atmosphere. Both processes do not allow the recovery of organic component of the plastic waste which could be part of organic life-cycle. Feed stock recycling of plastics waste is for management of plastic wastes. In such processes, plastics are transformed to their constituent monomers and or basic hydrocarbon [5].

The various types of recycling methods are good options to control the increase of plastic waste, because they are environmentally friendly when compared with incineration and disposal in landfills. Recycling is a possible way to recover useful raw materials, energy and fuel, while minimizing the consumption of natural resources and raw materials when these products of industrial activities are reduced [6].

The current plastic reclamation technology options are generally grouped into four types: Primary recycling, secondary or mechanical recycling, feedstock or chemical recycling and quaternary or energy recycling, [7]. The most popular process is represented by the primary recycling due to their simplicity and low cost. This is the processing of polyethylene for use comparable to the original application. The disadvantage of this method is represented in the existence of a limit on the number of cycles for each material [8]. The secondary process is represented by a physical method in which the polyethylene is shredded to granules and melted to make new products by extrusion. The processed material can be blended with virgin material to obtain superior results. The disadvantages of this method refer to the heterogeneity of the solid waste and the deterioration of product's properties in each cycles which occurs due to the low molecular weight of the recycled resin. This method is also relatively inexpensive but needs substantial initial capital investment [9].

Feedstock or chemical recycling which is also known as tertiary recycling is defined as the process in which polymers are chemically converted to monomers or partially depolymerised to oligomers through a chemical reaction. A change occurs in the structure of the

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polymer. The resulting monomers can be used for new polymerizations to produce the original or related polymeric product as well as recover gasoline range fuel, [10].

The chemical reactions used for decomposition Almeidf polymers into monomers are: Hydrogenation, glycolysis, hydrolysis, gasification, methanolysis, chemical depolymerisation, thermal cracking, catalytic cracking and reforming, photo degradation, ultrasound degradation, degradation in microwave reaction [11]. The chemical recycling is not fully developed and for this reason, only a few companies are working on it because this method is needs a lot of investment and expert personnel. Contributing to this high cost is the fact that waste polyethylene are indeed a mixture of different materials having different processing compositions and requiring different processing conditions. Another prevalent issue with this method is the high energy input required as some waste require as high as 700 °C. The quaternary or energy recycling method refers to recovery of plastic's energy content. The most effective way to reduce the volume of organic materials which involves the recovery of energy is represented by incineration. This method is a good method because it generates considerable energy from polymers, but it's not ecologically acceptable because of health risk from airborne toxic substances [13].

Thermal or catalytic cracking of plastic waste yields a mixture of basic hydrocarbons which are valuable either as fuels or as industrial raw materials. The energy consumption during the thermal cracking (pyrolysis) is very high and the molecular weight is broad, varying with the conditions used [14]. However, the selectivity of the product can be controlled by the use of a suitable catalyst (catalytic cracking) under appropriate conditions of temperature, pressure, and atmosphere (N_2, H_2 or air). Catalytic cracking of plastics requires lower energy consumption temperatures and the chemical distribution of products is narrower than in the thermal cracking processes leading to the production of more valuable products [15]. In this study, tertiary recycling of postconsumer waste low density polyethylene was considered and the pyrolysis reaction of the LDPE with and without the zeolite catalyst was carried out varying the temperature and the catalyst/sample ratio in order to recover combustible the gases evolved

EXPERIMENTAL

LDPE sample collection and preparation

The low density polyethylene waste (LDPE) mainly waste drinking water sachets were collected from refuse dumping sites around Makurdi town, Benue State of Nigeria. The samples were washed thoroughly using detergent and rinsed properly with deionized water, then dried at room temperature to remove all moisture. The dried samples were reduced into smaller bits to increase the surface area as described by [16]. The zeolite catalyst (X-23) employed for this work was a Sigma Aldrich Company product and is shown by analysis to be composed of aluminum and silicon. Scanning Election Microscopy (SEM) images of the zeolite catalyst were obtained at magnifications of 1000 x, 2500 x, 4000 x, 7500 x, 10 000 x and 15 000 x respectively. From the SEM images, the zeolite catalyst particle sizes range within 5 µm – 80 µm. In further characterizing the zeolite catalyst, SEM was combined with atomic absorption spectrometry (AAS) in the detection of the metal composition. The analysis indicated the presence of aluminum 6.3010 ppm_ and silicon 0.7220 ppm metals.

Experimental Procedures

A Presto pressure cooking pot made of stainless steel with dimensions of height 30.00 cm with an internal diameter of 31.50 cm was converted to a fixed bed reactor and adapted for the pyrolysis reaction [17]. The cover had an outlet tube at the top for collection of the evolved gases. The reactor was effectively lagged with a fire blanket and placed in sand bath constructed with iron sheets for good heat retention. The reactor fitted in the sand bath was heated with three Mekker burners in order to attain the required temperatures which were controlled by means of a thermocouple. Tedlar bags were used for collection of the evolved gases. These gases were immediately sent for gas chromatographic analyses.

2.2.1. Low Temperature Thermal cracking of LDPE

1.5 kg of (LDPE) samples were shredded and stuffed in the reactor and heated for about 3 hours until the temperature reached 350 ºC. The reaction temperature was monitored and controlled by means of a thermocouple. The gases started evolving after 1½ hours into the reaction and this was evident from the tedlar gas sampling bags which began to swell (at a temperature of about 120 ºC), the gas samples were collected at 200 ºC and 350 ºC and after the evolution of gases, the system was allowed to cool before the reactor was opened. The residue in the reactor was collected and the weight noted. The gases which were collected in a labeled tedlar gas sampling bags were sent for gas chromatographic analyses [18].

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2.2.2. Catalytic Low Temperature Cracking of LDPE

200 g of the LDPE sample was heated with the effect of the catalyst. Two catalyst/sample ratios were investigated with slight modifications [19]. In the catalytic reaction, the sample (polymer material) was placed at the bottom while the catalyst material was arranged in sandwich layers within the polymer sample [20]. The reactor was covered and then heating commensed. After 30 minutes into the reaction (at 120 ºC) evolution of gases started slightly and after 1½ hours at about 250 ºC the system was shut down. The gases which were collected at 150 ºC and 200 ºC in labeled tedlar gas sampling bags were then analyzed. The system was allowed to cool down before the reactor was opened. The residue after each run was collected and weighed.

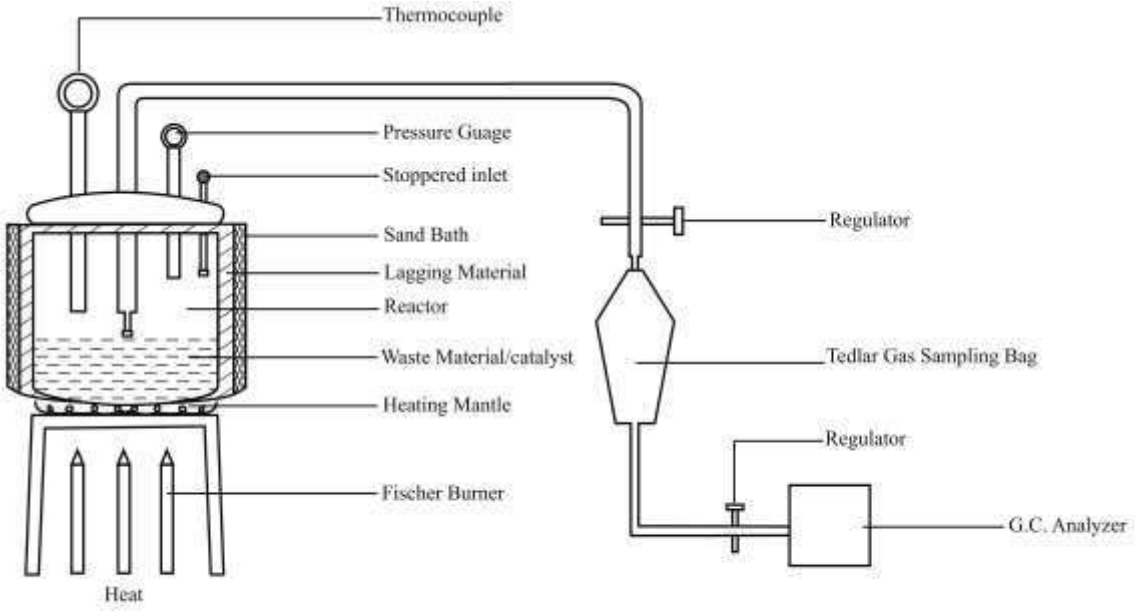

Fig. 1. Layout of the pyrolysis process

Gas Chromatographic Analyses

The weight obtained at the end of the thermal pyrolysis reaction was 850 g while 120 g and 130 g were obtained at the end of the catalytic pyrolysis at catalyst/sample ratios of 1:8 and 1:16 respectively.

The gaseous products obtained from pyrolysis of LDPE with and without the effect of catalysts were characterized using a Buck 530 Gas chromatograph and HP-88 columns. Injection volume was 10 µl with helium as the mobile phase and flame ionization detector (FID), injection and detection temperatures were 250 ºC and 280 ºC respectively.

RESULTS AND DISCUSSIONS

The gas chromatographic analysis of the gas sample obtained from the waste LDPE showed that the components present were mostly aliphatic in nature within the range of $C_1 – C_9$. Composition of gases obtained from pyrolysis of waste LDPE at 200 and 350 ºC is present in Fig 2.

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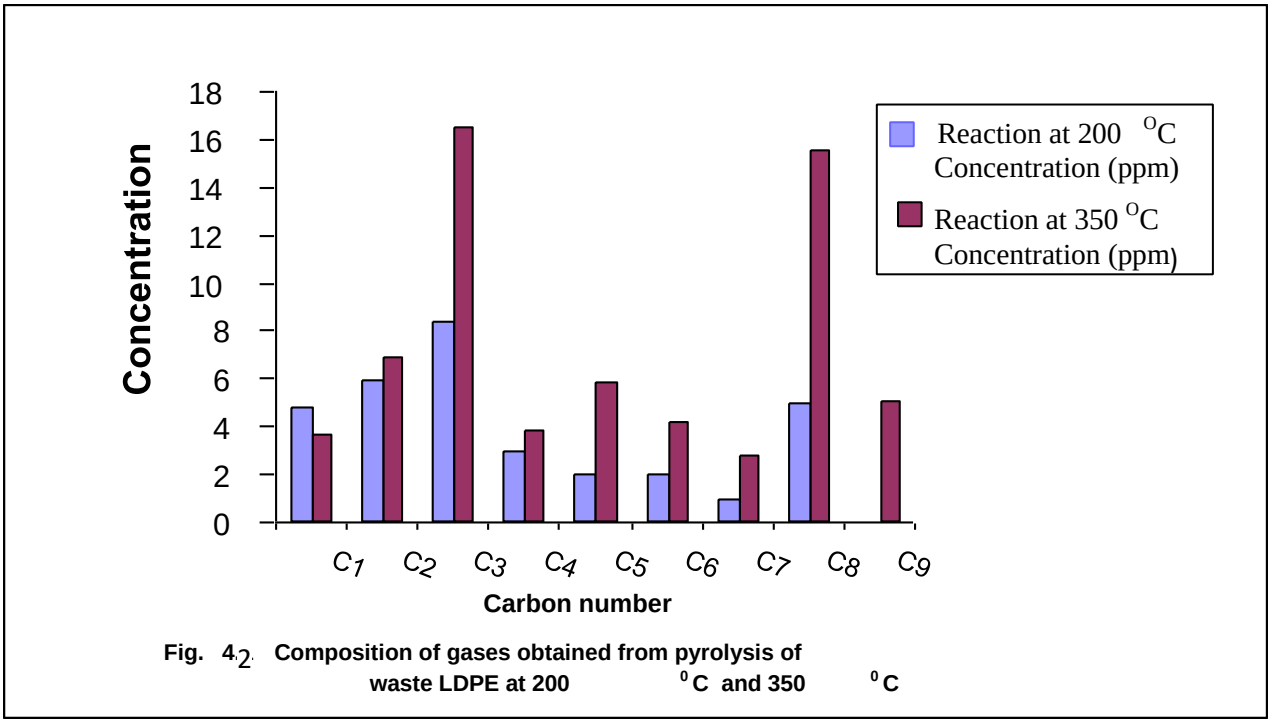

Fig. 4.2. Composition of gases obtained from pyrolysis of waste LDPE at 200 ^{0}C and 350 ^{0}C

How LDPE is converted to low molecular weight gases during pyrolysis is still a matter of conjuncture. Lost of a hydrogen atom followed by hemolytic β - carbon bond fission could be a plausible pathway. [21]

Waste sachets (LDPE) were pyrolysed at 200 ºC and a total concentration of 36.0906 ppm was obtained. The gases were in the range of carbon number C_1 – C_9. The dominant components were observed in C_1 4.7779 ppm; C_2 5.9438 ppm; C_3 8.3578 ppm; and C_8 4.9950 ppm. Water sachets were also pyrolyzed at 350 ºC also produced hydrocarbon gases. The total concentration obtained was 64.5041 ppm with the highest percentage concentrations in the carbon numbers C_2 6.9092 ppm; C_3 16.5089 ppm. This implies that the gaseous products obtained during the formation of wax from sachets are very useful; C_2 is one of the dominant components that was observed. This could be explained with the structure and degradation pattern of polyethylene because it is built up from C_2 monomers [22].

The yield of the gaseous products increased with temperature and time from 200 ºC – 350 ºC in Fig 2. This increase in gas yield could probably be due to the increase in the rate of main chain sigma cleavage reactions which are more thermally energetic within high temperature environments. The effect of reaction time on gas yield monitored up to 180 minutes. For LDPE, at about 90 minutes into the reaction, gases began to evolve and over time, the gaseous products increased, Thus 120 minutes into the reaction, the gases obtained were more than what had earlier evolved. This shows that the overall evolution and concentration of gases were more at higher temperatures (350 ºC), than what was obtained at slightly lower temperature. This agrees with the results obtained by Demibras and Ahyam, 2004 [23] indicating that higher yields are obtained at higher temperatures.

Waste water sachets (LDPE) were pyrolyzed at 150 ºC under the effect of zeolite catalyst in ratio of 1:8. The result of the gas chromatographic analyses is represented in Fig 3.

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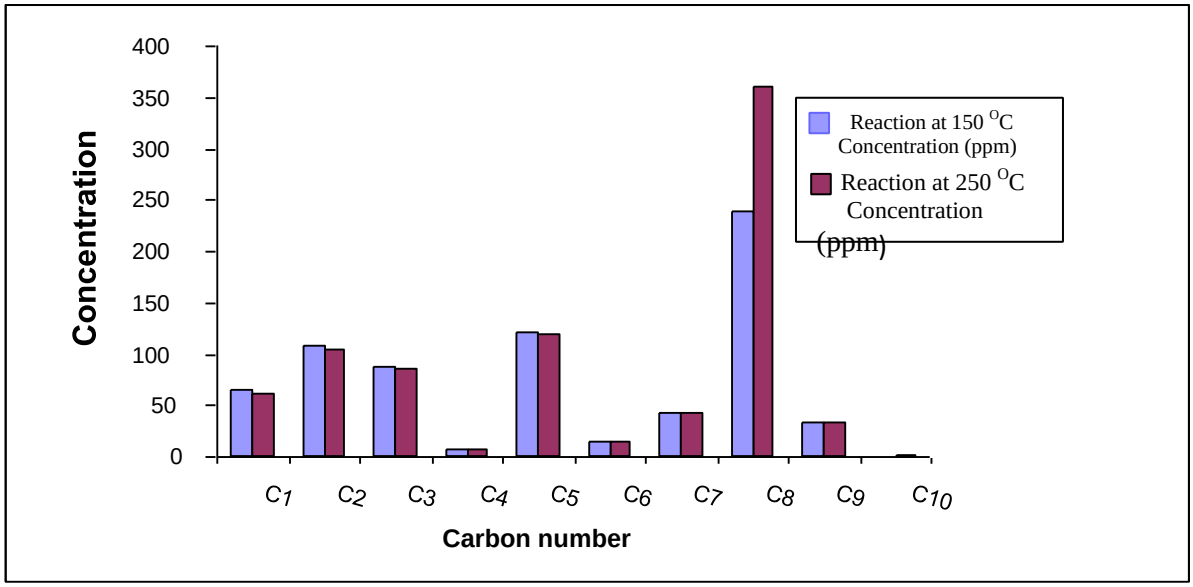

Fig. 3: composition of gases obtained from catalytic pyrolysis of waste LDPE at 150 ^{0}C at catalyst sample ratio of 1:8

From the result, the total concentration of gases obtained from its reaction at 150 $^{\circ}$C was 721.0371 ppm while the total concentration of gases obtained at 250 ºC yielded 835.0906 ppm. This implies that the gaseous products increased with increase in temperature from 150 ºC to 250 ºC. However, the total concentration of gases under the influence of zeolite catalyst were much higher than when the reaction occurred thermally even though the reaction took place at low temperature. This agrees with the fact that catalytic degradation of polymer has shown greater potential than purely thermal pyrolysis since it significantly lowers the pyrolysis temperature. This agrees with results presented by Murata *et al*, 2004 and Okhita,*et al*, 1993, [24-25].

The most prominent peaks from the result were observed in C_1 65.6505 ppm, C_2 108.1654 ppm, C_3 87.0287 ppm and C_5 238.9797 ppm. It was also noted that in the reaction carried out at 150 ºC, the presence of C_{10}
0.8487 ppm was observed. The result of the gas sample analysis obtained at 250 ºC revealed that the dominant carbon atoms were C_1 62.0894 ppm, C_2 105.0067 ppm, C_3 86.8908 ppm, C_5 120.0705 ppm, C_8 361.6030 ppm. Also, the concentration of C_9 33.9074 ppm increased considerably with C_{10} 1.1113 ppm carbon atom also increasing in concentration.
This result shows that the lighter hydrocarbons increased in concentration over what was obtained thermally. This is in accordance with the expectation that the catalyst should enable breaking down of the polymer feed to lighter yield when compared with the reaction which occurred without the catalyst [26]. It is widely observed that zeolite catalysts would favour the production of gases in fuel feed stock recycling. This is also evident in this result as the concentration of gases has increased greatly compared with the sample reacted without the catalyst. This high gaseous products could be attributed to the over cracking nature of the zeolite which are solid acids [27]. In addition, significantly lighter hydrocarbons were formed.
Similarly, results obtained from pyrolysis of LDPE at 150 ºC and 250 ºC using catalyst/sample ratio of 1:16 gave a total concentration of gases at 150 ºC and 250 ºC to be 697.4464 ppm and 713.7277 ppm respectively.

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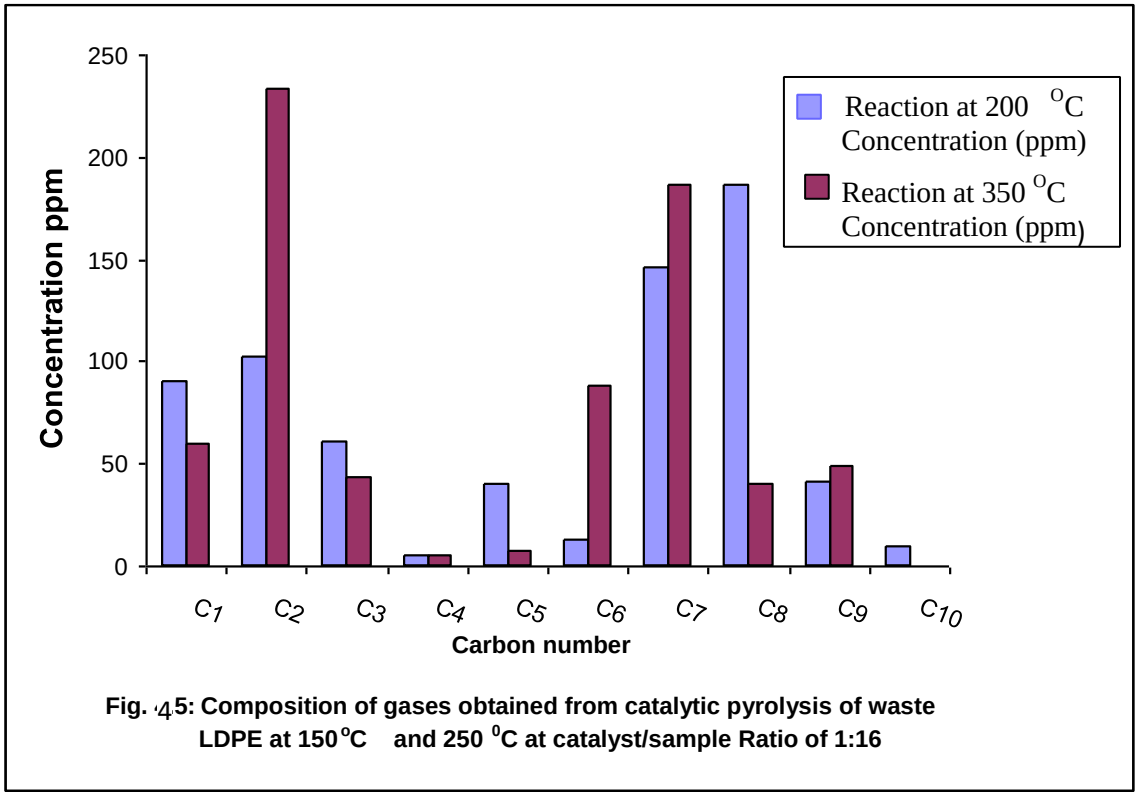

Fig. 45: Composition of gases obtained from catalytic pyrolysis of waste LDPE at 150°C and 250 °C at catalyst/sample Ratio of 1:16

Fig 4: Composition of gases obtained from catalytic pyrolysis of waste LDPE at 150 and 250 °C at catalyst/sample ratio of 1:16

This is an indication that the concentrations increased with increase in temperature. The total gas yield under the influence of zeolite catalyst was more in concentration than when the reaction occurred only thermally. This agrees with results presented by Surma, et al, 2020 [28]. Based on these results it can be deduced that the zeolite catalyst results in very high gas yields in the range of $C_1 – C_9$ products and apparently in much higher concentration than observed in the thermal cracking reactions. These zeolite particles were this effective because of their surface area. This agrees with results obtained by Aguado et al, 2004 [29] when they studied the influence of the operating variables on the catalytic conversion of polyolefin mixture over HZ5M-5 zeolite catalyst and obtained high gas yield in the range of $C_3 – C_5$ products. It is also observed from these reactions that conversion increase with catalyst rating as the total concentration of gases obtained using catalyst/sample ratio of 1:8 was higher than that of 1:16. This agrees with works done by Akpandoh *et al* (2005) [30].

CONCLUSION

Combustible gases were recovered from thermo-catalytic pyrolysis of low density polyethylene. This study has therefore demonstrated that low temperature pyrolysis of low density polyethylene whether catalyzed or uncatalyzed is effective method that could use to recover reasonable conversion of waste polyethylene materials to useful gases and at the same time reduces the bulk size of the waste. The thermal cracking reaction yielded fuel gases mainly within the range of $C_1 – C_{10.}$ The zeolite catalyst has shown high activity for hydrocarbon cracking reactions with selectivity for aliphatic hydrocarbons within the range of (C_1- C_4 and C_5-C_{10}) and (C_1C_4 and $C_5 –C_6$) for the reactions which occurred using catalyst/sample ratio of 1:8 and 1:16 respectively. This method if well developed it will serve as a viable method in controlling environmental and resources problems perpetuated by plastic waste disposal and therefore, there will be conservation of resources.

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