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PRODUCTION OF SYNTHETIC FUELS FROM HIGH DENSITY POLYETHYLENE (HDPE) WASTE THROUGH PYROLYSIS: EXPERIMENTAL AND SIMULATION APPROACHES

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Abstract: Production of synthetic fuels through the pyrolysis of high density polyethylene (HDPE) waste was examined. A semi-batch reactor was designed and fabricated to pyrolyse HDPE waste in the absence of catalyst at a residence time of 60 minutes and final temperature of 425°C. At the end of the process, 51.84%, 45.33%, and 2.83%, liquid product, char and evolved gaseous product were obtained respectively. The temperature progression for the process was examined. The pyrolysis oil obtained was light brown in colour, highly flammable, with a density of 772.6 kg/m³ and pH of 5.5. ASPEN Hysys was used to simulate the pyrolysis process. Simulation results revealed an oil yield of 97%, a gas yield of about 2% and a <1% char yield. The simulation revealed the commencement of reaction at about 325°C with an optimum reaction temperature of 450°C. If properly optimized HDPE is revealed to be an excellent feedstock of pyrolysis.

Keywords: Pyrolysis, HDPE, Synthetic fuels, ASPEN Hysys, Simulation

1. INTRODUCTION

Due to economic growth and changing consumption and production patterns, there has been a rapid increase in the generation of waste plastic in the world. In the United States for example, municipal waste generation rose from 88.1 million tons in 1960 to 250.9 million tons in 2012 [1]. These figures rise at a rate of 5% yearly [2]. The increase in generation has led to plastics waste becoming a major stream in solid waste. After food waste and paper waste, plastic waste is the major constituent of municipal and industrial waste in cities [3]. Even cities with low economic growth have started producing more plastic waste due to an explosion of the applicability of the different types of plastics. This increase has now led to a major challenge for the authorities responsible for waste management. European Union (EU) has already mandated that by 2020, all plastic waste must go to mechanical, thermal or chemical processing facilities. No more waste will be allowed in landfills [2].

In countries where there is a lack of integrated solid waste management, most of the plastic waste is neither collected properly nor disposed in appropriate manner to avoid its negative impacts on the environment and public health. Landfill and incineration are two common means of plastic waste disposal which are currently being used which have severe impact on the environment [4]. 60% of all plastic solid waste currently goes to landfill [2]. Landfill could result to plastic additives such as phthalates and various dyes polluting ground water. Incineration results in the formation of unacceptable emissions of gases such as nitrous oxide, Sulphur oxides, dusts, dioxins and other toxins. On the other hand, plastic waste recycling can provide an opportunity to collect and dispose plastic waste in the most environmentally friendly way and it can be converted into a resource.

Recycling of plastic already occurs on a wide scale. Extensive recycling and reprocessing of plastics are performed on homogenous and contaminant free plastic wastes into new plastic products with a lower level of quality. The disadvantage of such an approach is that the product no longer has the special characteristics of the plastics used to make it. Thus, the product is less useful and end up competing for markets with cheap construction materials. The presence of non-plastic contaminants leads to concern from potential buyers about product quality and consistency. For this reasons, mechanical recycling of mixed plastic waste appears to have only a limited future. Tertiary recycling returns plastic to their constituent monomers or to a higher value hydrocarbon feedstock and fuel oil. Tertiary recycling includes all those processes which attempt to convert the plastic wastes to basic chemicals by the use of chemical reactions such as hydrolysis, methanolysis and ammonolysis for condensation polymers, and to fuels with conventional refinery processes such as pyrolysis, gasification, hydro-cracking catalytic cracking, coking and vis-breaking.

Pyrolysis and catalytic conversion of plastics is a superior method of reusing the waste. The distillate product (pyrolysis oil) is an excellent fuel and makes the process one of the best, economically feasible and environmentally sensitive recycling systems in the world today. In the turn of the millennium, there were as much as 100 small scale pilot plants globally processing more than 4000 tonnes of residue per year [5]. Pyrolysis plants are typically used to degrade carbon rich organic and inorganic materials such as biomass, municipal and industrial waste.

A lot of work has already been done as regarding pyrolysis technology as it applies to plastic materials. Low, Connor [6] described a simple pyrolysis reactor system, results of their pyrolysis tests showed that pure samples of polyolefinic and polystyrenic resins can easily be pyrolysed to produce liquid yields in excess of 70%. Feng [7] optimized the processes of

plastic pyrolysis for maximizing the diesel range products and designed a continuous pyrolysis apparatus as a semi-scale commercial plant. Caruso William, Danielle Sorenson [5] examined the viability of plastic pyrolysis as an alternative energy technology that will serve as a solution for urban carbon reduction. They concluded that it is quite feasible and has good prospects. Thorat, Sandhya Warulkar [8] prepared a report to show that pyrolysis oil is a truly sustainable waste solution utilizing the embodied energy content of plastics and producing a highly usable commodity. Recently, Guarav, Madhukar [3] pyrolysed low density polyethylene to get fuel oil at a yield of about 70% of the product with a simple reactor design. As regards high density polyethylene (HDPE) in particular, studies has been carried out to understand the kinetics [9-14], yield [15, 16], characteristics [10, 11, 17] and simulation [18, 19] of the pyrolysis process.

The aim of this research is to investigate the yield of high density polyethylene waste as feedstock for the production of synthetic fuels via the pyrolysis process with a close investigation on the effect of some subtle factors like the reactor design and temperature progression. A predictive simulation model using ASPEN Hysys 2006 was also developed for the process and was used for a sensitivity analysis and a reference system in predicting product yield and process behaviour/response to operating factors. This study comes at a time when concerted effort is made worldwide to curtail the amount of plastic waste going to landfill and incineration. These processes have been considered harmful to the environment. Plastic waste generation has been on the rise over the past few decades as more applications of plastics are being explored. This has led to the current status quo of which, numerous processes for recycling these wastes are currently investigated and optimized. Pyrolysis is one of the most recent and promising recycling techniques that researchers have come up with. Any investigation into the pyrolysis process such as this is by all means relevant as it would have contributed to the cause of developing and optimizing current recycling techniques for plastic waste.

2. METHODOLOGY

— Design of the semi-batch reactor

The first stage was the design and fabrication of a miniature semi-batch plastic pyrolysis reactor for the purpose of this study. A schematic diagram of the design is presented in Figure 1 while the specification sheet for the reactor is presented in Table 1.

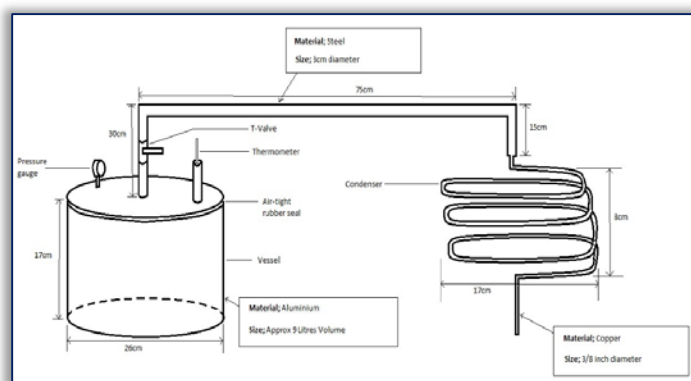


Table 1: The specification sheet for the reactor

Table 1. The specification sheet for the reactor

Identification: Semi-Batch Plastic Pyrolysis Reactor		By: Adeniyi, A.G., Osemwengie S. O. and Ighalo Joshua O	
Function; Non-catalytic pyrolysis of high and low density polyethylene (HDPE & LDPE)			
Operation; Semi-batch			
Design Data			
Reactor vessel Material Handled; HDPE Temperature; $\leq 450^{\circ}\text{C}$ Material of construction; Aluminum Pressure; 2 bars	Shape; Cylindrical body with a tori-spherical top Diameter; 26cm Height; 17cm 1 product escape route		
Condenser Materials handled: Product vapour Temperature; $\leq 450^{\circ}\text{C}$ Material of construction: Copper	Pipe diameter; 3/8in 3 rings 8cm wide 17cm high		
Product escape Pipe Material of construction: Steel Material handled; Product vapour	Length; 75cm Pipe diameter; 3cm		
Peripherals: Globe valve, pressure gauge, thermometer & cork, Nitrogen Additional information: Personal protective equipment (PPE) includes Chemical respirator and high temperature gloves			

— Method for the Experiment

The second step was to source for samples of high density polyethylene (HDPE) present in municipal waste and practically pyrolysing them. Samples of high density polyethylene (HDPE) in the form of jerry cans was picked from different dump sites of municipal plastic waste in University of Benin, Benin-city, Edo State. The HDPE jerry cans were broken into large pieces and then thoroughly washed with detergent and water to remove dirt. They were afterwards rinsed with clean water. The HDPE samples were sundried from one (1) day. They were then cut into smaller sizes of about 1mm by 1mm. This was done to conserve the effective density of the plastic and to ensure heat flow through the sample at the initial stage of heating. The samples were then weighed with the Mettler PM4800 electric weighing balance.

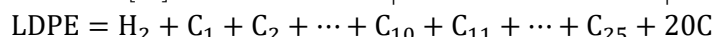
The reacting vessel was cleaned and 1kg of high density polyethylene (HDPE) samples was introduced. The reactor was sealed. With the product escape valve open, a stream of gaseous nitrogen was introduced into the system for 60 seconds. This was to purge out the oxygen present in the reactor and provide an inert environment for pyrolysis to take place. The product escape valve was closed and the thermometer and cork for temperature measurement was introduced. The entire

set up (of the reactor) was heated by an LPG burner. The rate of heating of the reactor was approximately 7°C/min and constant monitoring of the process was ensured. The product escape valve was opened at 270°C though pyrolysis commences at a higher temperature, this was done to avoid any pressure buildup within the reactor. A slight positive pressure was noticed throughout the experiment. The experiment lasted for one (1) hour after no significant formation of product was noticed signaling the end of the pyrolysis process.

— Method for the Simulation Study

ASPEN Hysys 2006 was used in modelling the pyrolysis of high density polyethylene. The reaction stoichiometry can be used to represent the reaction sequence. The sequence is represented in Figure 2.

As observed from the sequence, secondary reactions are considered to be absent as the process is approximated as a direct conversion of the feedstock to the respective products. The equation of reaction utilized is chosen in such a way that the above sequence can be easily represented. The equation is presented below and is a modification of that of Alla and Ali [19]. The reaction was stipulated to occur in the vapour phase alone.



The kinetics of waste HDPE has been extensively studied by numerous researchers [9-14]. For this study, the kinetic parameters for waste HDPE pyrolysis obtained by Kayacan and Doğan [14] at a heating rate of 5 K/min was utilized. The reported values are $A = 3.42 \text{ E}27 \text{ s}^{-1}$ and $E = 420.86 \text{ KJ/mol}$. The Arrhenius rate equation is given by the expression in equation 1

$$K = A \text{ Exp}\left(-\frac{E}{RT}\right) \quad (1)$$

where $K \text{ (s}^{-1}\text{)}$ is the rate constant, $A \text{ (s}^{-1}\text{)}$ is the pre-exponential factor, $E \text{ (KJ/mol)}$ is the activation energy, $T \text{ (K)}$ is temperature and R is the universal gas constant 8.314 KJ/molK .

The Peng-Robinson (PR) property package was used in the simulation. The databank in Aspen HYSYS 2006 does not contain any polymers. The method used was to represent the HDPE polymer feed as a hypothetical component. The chemical species used to model the feed is ethylene. This information informs the software of the elemental composition of the feedstock. However, to completely predict all other physical and chemical properties of the feedstock, Aspen HYSYS requires the stipulation of three properties; density, molecular weight and normal boiling point. These properties were inputted into the hypothetical component manager and they served as the basis with which the software estimated the other necessary information about the polymer. The properties of HDPE inputted into the hypothetical component manager includes a Density of 940 kg/m^3 [20], Molecular weight of 46200 g/mol [20], Normal boiling point of 270°C (predicted from experiment) and Normal melting point of 125°C (predicted from experiment). Other reaction products (H_2 , carbon, $\text{C}_1\text{-C}_{24}$) were added as conventional components in the simulation. The process is represented in Figure 3.

The feed (1kg/hr) is sent into the reactor at ambient conditions (25°C) and 1atm . The continuous stirred tank reactor (CSTR) was chosen as the pyrolyser in the simulation as the reaction kinetics and stoichiometry could be easily implemented on it. The pyrolysis temperature was set 425°C . The bottom product from the reactor was removed as the char stream while the vapour stream was sent to a condenser unit operating at 25°C and ambient pressure. The non-condensable gases leave the condenser as top product synthesis gas stream while the condensate pyrolysis oil leave in a bottom stream. Several assumptions were taken in developing the simulation model. The pyrolysis reaction was considered to take place only in the vapour phase. The process was studied at steady state hence time is not included as a factor. The char was assumed to be composed solely of elemental carbon. In practical scenarios, bio-char from pyrolysis sometimes contain heavy metals but this will not be implemented in the simulation

3. RESULTS AND DISCUSSION

Considering the Table 1, it can be observed that about half the high density polyethylene sample was converted to liquid product. Just over 45% was residual char while the rest was gaseous product. Looking at this result, it must be said that the amount of char produced by the process is worryingly large. This is due to the design intricacies of the reactor. When

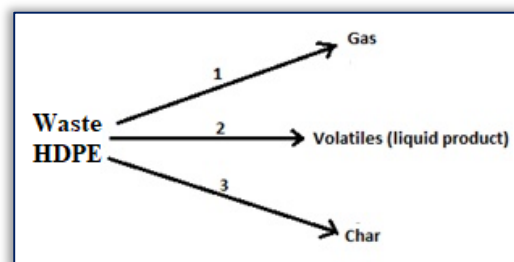


Figure 2: The Reaction Stoichiometry Sequence

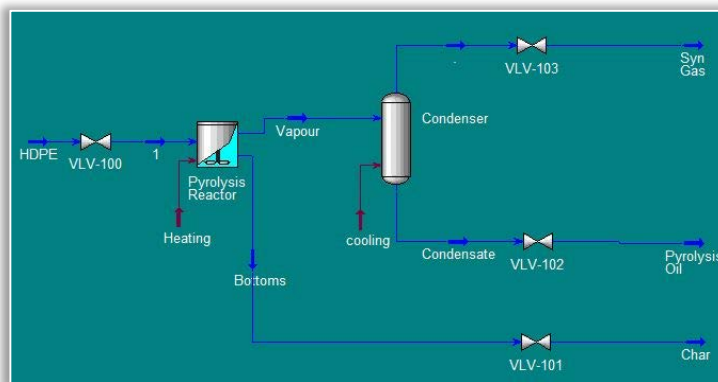


Figure 3: The Pyrolysis of HDPE Waste

the heating rate is not rapid and product elutriation is not optimal due to reactor shape, the pyrolysis process tends to be slow hence favouring higher char formation. Several other factors may likely have contributed to this but a clearer picture will be obtained when these results are juxtaposed with those of other researchers.

Low, Connor [6] pyrolysed High Density Polyethylene (HDPE) using a semi-batch laboratory apparatus setup at a temperature of about 400°C for 80 minutes. They obtained 70% liquid product, 28% gas and 2% residue. Residue obtained was characteristically low too. Though both experiments are similar in terms of catalysis, residence time and temperature, the type of reactor used are quite dissimilar. An experimental laboratory setup will always perform higher than larger scale setup. There is minimal temperature variation in an experimental apparatus compared to larger scale setup. Also, the removal of product from the reaction chamber is more efficient as it approximates ideality far more than larger scale setup. A deviation from ideality usually occurs in reactors in the form of stagnant regions, short-circuiting and channeling [21]. Feng [7] pyrolysed virgin HDPE in a batch apparatus at an end temperature of about 800°C in an experiment lasting for 60 minutes. He obtained 83.5% liquid product, 0.5% char and 16% gaseous product. The result of this experiment is quite similar to that obtained by Low, Connor [6] which displays a fantastic amount of liquid product yield and very little amount of left over residue. The results of Williams and Williams [15] were also quite similar to those of Feng [7] albeit at a heating rate of 25°C/min and a final temperature of 700°C. We notice that pyrolysis char formation can be minimized by utilizing very high temperatures for the process. However, considering the energy requirement of such a process, economic considerations should be considered and low temperature optimization of the process becomes a viable option.

The result of Achilias, Roupakias [16] (Table 3) for waste HDPE at 450°C is fairly consistent with that of the current study. Though utilizing a fixed bed reactor, their process was catalytic (FCC) and the setup was laboratory scale. There was a constant stream of nitrogen purge gas has product elutriation was faster. Reported residence time was 17 minutes. Alla and Ali [19] developed a simple simulation model for polyethylene pyrolysis and for a temperature of 450°C obtained 95.2% oil, 4.6% gas and <1% char. A different and more detailed and predictive model was developed in this study.

Table 3: Comparison of this Work with Previous Work

Product	Current study	Achilias et al. (2007)	Low et al. (2001)	Feng (2010)	Williams and Williams (1997)
Temp	425°C	450°C	400°C	800°C	700°C
Char	45.33%	52.50%	2%	0.50%	0.00%
Liquid	51.84%	44.20%	70%	83.50%	79.72%
Gas	2.83%	3.30%	28%	16%	16.77%

— Temperature-Progression Profile for HDPE Pyrolysis

After the first 2-3 minutes of heating, no significant change occurred in the reactor. The metal of the reactor body is still heating up and little energy reaches the polymer. Subsequently, the temperature of the polymer rose at a steady rate (approx. 7°C/min) for about 13 minutes till the 125°C threshold was attained. There was an interesting thermal behavior at this point because upon heating, little or no change occurred in the temperature of the system for a while. This was due to the fact that the polymer samples melted somewhere within 125°C to 130°C. The latent heat of fusion makes it impossible for the temperature to climb, as extra work needs to be done (in the form of heat) to change the state of matter from solid to liquid. Beyond this temperature, a steady rise was re-established and similar to the pre-melting point temperature rise of about 7°C/min. Once again, after about 30 minutes, the lag was observed again. It occurred at about 270°C. This is also likely to be the vapourisation temperature. At temperatures beyond 270°C, a lot began to happen within short time intervals. As the temperature rose, the initial stage of pyrolysis commenced. The long-chain polymers break into compounds of shorter chain length and the first pyrolysis products was evolved. The initial liquid product appeared to be waxy because the polymer chains in the product are still quite long as only primary pyrolysis has taken place. Beyond 350°C, the temperature climbs very rapidly (100°C in about 10 minutes). It is during this period that the

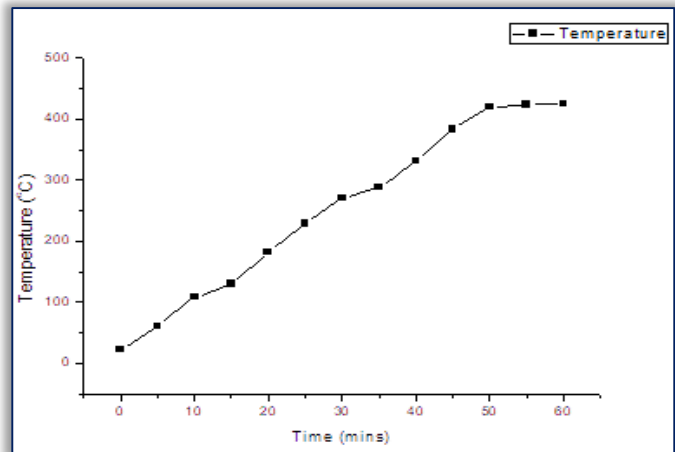


Figure 4: Temperature-Progression Profile for HDPE Pyrolysis

Table 2: Product yield from the pyrolysis experiment

Product	Yield
Char	45.33%
Liquid	51.84%
Gas	2.83%
Pyrolysis temperature; 425°C, Residence time; 1 hour	

main bulk of the product was collected. The oil produced during this period was rather clear. Beyond 400°C, there was no longer any significant rise in the temperature of the reactor with time. Most of the products have being collected and only a little amount of the oil was being evolved. The process was finally stopped at about 425°C when it was noticed that no significant amount of oil and synthesis gas was being produced by the system.

Feng [7] also monitored the temperature profile of plastic pyrolysis albeit with mixed polymer samples. Though allowing his experiment to run for about 300 minutes, temperature rise only occurred approximately within the first 80 minutes of reaction, and he obtained a profile quite similar to the one obtained in this research work. However, due to his experimental methodology and other factors (such as heating rate), the latent heats (of fusion and of vapourisation) were not noticed. However, Low, Connor [6] using a heating rate of 33°C/min for a residence time of 80 minutes obtained similar results. Several factors affect the nature of temperature progression in pyrolysis reactions of paramount significance is the reactor type. The design intricacies of the reactor will determine where the temperature readings will be taken from. It can be from the liquid, the vapour, the inner wall of the reactor, reflux inlet or outlet etc. Another factor is the starting polymer material; different polymers will pyrolyse at different temperature ranges. Heating rate and the presence of catalyst are also important factors.

— Basic Properties of the Pyrolysis Oil

The pyrolysis oil obtained this research work was observed to be of light brown colour. This result (Table 4) is consistent other experimental results. It is highly flammable as expected of any combustion fuel and burns in air without any residue. The pH was found to be 5.5. Cole [22] noted that pyrolysis fuels are generally acidic hence tends to corrode most steel storage tanks, fuel lines and engine parts. The density of the pyrolysis oil obtained is 772.6kg/m³. Guarav, Madhukar [3] obtained a liquid product with a density of 702.5 kg/m³ and Low, Connor [6] obtained a liquid product with a density of 750 – 800 kg/m³ range. The density of conventional diesel is 830kg/m³. The density obtained in this research work is consistent with those obtained by other researchers. As revealed by Wongkhorsub and Chindaprasert [23] this oil can serve as a suitable substitute to diesel, albeit at a lower performance level and higher production cost.

— Simulation results

The results in Table 5 show the oil yield of HDPE pyrolysis from the simulation. Results of Alla and Ali [19] for polyethylene pyrolysis is noticeably similar than those of the simulation. Simulation results will always demonstrate above average oil yields because they are essentially idealized and many of the extraneous factors obtainable in conventional experiments cannot be incorporated into these softwares. Accuracies are also determined by the chosen reaction scheme, kinetics and approximate stoichiometry. These results however prove that if properly optimized, high density polyethylene is an excellent feedstock for pyrolysis will give very good yield of bio-oil.

The reaction conversion from the CSTR was monitored with the temperature of reaction to obtain the plot given in Figure. It reveals the commencement of HDPE pyrolysis at about 325°C and optimal reaction conversion at 450°C. From experiment, a majority of the products was evolved from the system between 350°C and 400°C. We can surmise that the optimum reaction temperature for high density polyethylene pyrolysis is 450°C. The above profile is consistent with experimental results for obtained for other plastics by researchers [24, 25].

4. CONCLUSIONS

This research work is simply an attempt to pyrolyse high density polyethylene (HDPE) waste with an eye to obtain very valuable pyrolysis oil while simultaneously proffering solution to the malignant issue of plastic waste disposal. This aim was to be achieved by estimating the amount of pyrolysis oil obtained as well as examining the temperature progression for the process. A semi-batch plastic pyrolysis reactor was designed and fabricated for this purpose. HDPE samples were pyrolysed in the absence of catalyst at a final temperature of 425°C and a residence time of 60 minutes. 51.84% of liquid product was obtained, 45.33% char remained in the reactor while the remaining 2.83% was gaseous product. The pyrolysis oil obtained was a light brown highly flammable liquid with a density of 772.6 kg/m³ and a pH of 5.5. ASPEN Hysys was used to simulate the pyrolysis process. The reactor was modelled by a continuous stirred tank reactor requiring both stoichiometric and kinetic information. The model functioned as a reference system in predicting product yield and

Table 4: Table Basic Properties of the Pyrolysis Oil

Property	Value
Colour	Light brown
Flammability	Highly Flammable. Burns without residue
pH	Acidic (5.5)
Density	772.6kg/m ³

Table 5: Oil yield of HDPE Pyrolysis from the Simulation

Products	Current Study	Alla and Ali (2014)
Temperature	450°C	450°C
Char	0.20%	<1%
Liquid	97.43%	95.20%
Gas	2.37%	4.60%

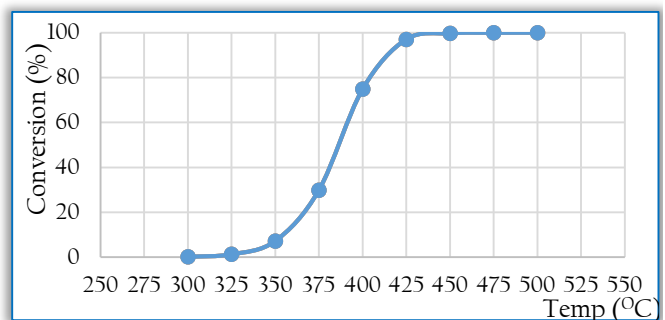


Figure 5: The reaction conversion profile

process behaviour/response to operating factors (such as temperature). Simulation results revealed a bio-oil yield of 97%, a gas yield of about 2% and a <1% char yield. The simulation revealed the commencement of reaction at about 325°C with an optimum reaction temperature of 450°C. If properly optimized high density polyethylene is revealed to be an excellent feedstock for the pyrolysis process.

Due to the fact that a good product yield is obtainable, waste plastic pyrolysis can be a lucrative venture. Entrepreneurs are hereby encouraged to explore this possibility. Investors should make concerted effort to construct large-scale pyrolysis plants (especially in the major cities where plastic wastes are readily available) to help deal with the issue of plastic waste disposal. A major issue noticed is a lack of market (worldwide) for pyrolysis oil because it is considered unstable and acidic [26]. Researchers should investigate means of modification and refinements of the oil so as to enable it compete favourably in the market.

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