

Thermogravimetric Analysis and Kinetic Study on the Pyrolysis of Some Post-Consumer Plastic Wastes

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ABSTRACT

The management of post-consumer plastic wastes via pyrolysis is a very promising technique to resolve the crises posed by plastic pollution on the environment. Products derived through the pyrolysis of plastic can be employed as fuel for the production of power and feedstock for chemical processes. Thermogravimetric study of three common post-consumer plastic wastes [Polystyrene (PS), Low-density Polyethylene (LDPE) and Polyethylene terephthalate (PET)] were carried out to investigate and derive necessary data on their decomposition and pyrolysis behaviour. Thermogravimetric analysis showed that the degradation of all plastic wastes occurred in a single stage. Among the samples considered, LDPE has the highest rate of mass loss. Kinetic study revealed LDPE has the highest activation energy (E_A) of 242.9 kJ/mol with PET having the least E_A of 189.05 kJ/mol. The E_A and regression coefficient (R^2) were determined using Coats-Redfern method and thermogravimetric analysis (TGA) data. All post-consumer plastic wastes considered in the study can be utilized for energy production via thermochemical processing as revealed from their decomposition behavior and pyrolysis kinetics.

1. INTRODUCTION

Plastics represent a very important product of our modern society. Plastic materials have inherent properties making them have superior performance both in daily life activities as well as industrial applications. There are diverse types of plastic produced annually around the world; however, findings have shown that the most common plastic wastes polymers are polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET) and polyvinyl chloride (PVC) (Gerassimidou *et al.*, 2020). The annual worldwide production of plastic in 2016 is estimated to be 335 million tons (Crippa *et al.*, 2019). The rise in global population and rapid economic growth has also increased the demand for plastic products resulting into a corresponding increase in the amount of plastic wastes generated. The management of plastic wastes is a challenge both in the developed and developing societies of the world (Koushal *et al.*, 2014). Studies have shown that over 70% of plastic produced since 1950 are out of service and have either been dumped in landfills or dumped directly into the environment (Jambeck *et al.*, 2015; Pan *et al.*, 2020). As a result, plastic products contribute significantly to the problem of environmental pollution since they take billions of years to degrade (Surenderan *et al.*, 2018).

The development of effective means of processing post-consumer plastic waste to improve the efficiency of its utilization is very important. So much attention has been placed on the recycling of plastics as an

alternative means of disposing and managing plastic wastes in recent times. The recycling of plastic waste is broadly classified into two main pathways, that is, the reprocessing via mechanical means and the breakdown of plastic wastes into diverse range of chemicals or fuels using heat and/or catalyst (Kple *et al.*, 2017). To mitigate the sorting challenge posed by the mechanical reprocessing of plastic wastes leading to the process being labour intensive, the conversion of plastic wastes via thermal degradation to chemicals and fuels represents a sustainable way to manage plastic wastes while simultaneously depleting the product of degradation to meet energy demands. Plastic are derived from petroleum and possess similar fuel and thermal characteristics to other petroleum-based products. Plastics materials can be converted into fuel using diverse methods by the application of thermal energy like pyrolysis, incineration and gasification (Moustakas and Loizidou, 2010). Among the various technologies available for the thermal conversion of plastics, pyrolysis is the most desirable because it leads to a considerable decrease in the volume of waste; results in the production of a wide array of fuel and chemicals from plastic; recovers more energy and requires lower decomposition temperature and cost (Patni *et al.*, 2013; Moustakas and Loizidou, 2010).

Pyrolysis represents a very promising technique for the recycling of plastic solid wastes and it involves the degradation of longer chain polymers molecules via thermal treatment into simple, smaller molecules in an inert atmosphere. Three main products are obtained during the pyrolysis of polymers vis pyrolysis oil, non-condensable gas and char which are utilized in many industries. Pyrolysis is capable of yielding a high volume of pyrolysis oil (80%wt) at a temperature of around 500°C (Fakhrhoseini and Dastanian, 2013). Pyrolysis liquid may be utilized directly as energy sources in different applications such as boilers, furnaces and turbines and it can also be upgraded into transportation fuel which can be used in diesel engines. Pyrolysis oil is environmentally friendly since it also contributes to the reduction of CO₂ into the atmosphere (Abnisa *et al.*, 2014).

Several researchers have investigated the utilization of thermogravimetric analysis to determine the thermal behavior of materials via pyrolysis (Oyedun *et al.*, 2014; Kple *et al.*, 2017; El Abbari *et al.*, 2019). Na *et al.* (2008) worked on the pyrolysis characteristics of different medical wastes. Aboulkas *et al.* (2010) investigated the thermal behavior and pyrolysis kinetics of different types of plastic wastes. Also, Nzioka *et al.* (2019) employed TGA in the determination of the thermal behavior of municipal solid waste. Oyedun *et al.* (2019) studied the thermal decomposition of polystyrene (PS) and High density polyethylene (HDPE) and biomass with their blends. They observed that pyrolysis behavior of plastic materials is characterized by one stage reaction while the thermal behavior of the blends is characterized by two stages of decomposition. In addition, many studies have been conducted by employing different isoconversional methods together with TGA. Pan *et al.* (2020) evaluated the pyrolysis kinetic parameters of pure polyethylene, waste polyethylene and waste plastic using genetic algorithm and isoconventional methods coupled with TGA.

To ensure environmental sustainability and proper management of post-consumer plastic wastes, the use of post-consumer plastic wastes for pyrolysis process is desirable. There is dearth of data on the decomposition behavior and kinetic parameters of post-consumer plastics in Nigeria and sub-Saharan Africa. Therefore, the thermal decomposition behavior of some common post-consumer plastic wastes has been studied using a thermogravimetric analyzer to obtain more information on their decomposition behavior and obtain the kinetic parameters of these materials during pyrolysis. This is with a view to optimize the efficiency of post-consumer plastic during conversion and utilization.

2. METHODOLOGY

2.1 Preparation of Samples

Three different waste post-consumer plastic samples [polyethylene terephthalate (PET), polystyrene (PS) and low density polyethylene (LDPE)] were collected from Ile-Ife, Osun State, Nigeria. LDPE is a tough, flexible and relatively transparent plastic material; this material is usually employed in packaging drinking water in 500 ml heat-sealed plastic sleeves commonly known as sachet water in Nigeria. PET is a strong

and stiff synthetic fiber belonging to the family of polyester of plastic and is commonly used as a disposable bottle for packaging water for commercial purposes in urban Nigeria. PS is a hard and transparent synthetic organic material used for packaging food materials. LDPE and PET waste samples were obtained from the production wastes of a commercial water treatment and packaging company while waste PS samples were obtained from empty food containers on a dump site. These samples were thoroughly cleaned and the plastic samples were then shredded to approximate equal sizes of 10 x10 mm and then dried in air (Yuliansyah *et al.*, 2015).

2.2 Proximate Analysis

The proximate analysis of each of the waste plastic samples were carried out to investigate the percentages of fixed carbon, volatile matter, ash and moisture content in each sample using various methods as shown in Table 1.

Table 1: Methods Employed for Proximate Analysis

Property	Analytical Method
Moisture Content (MC)	ASTM D871
Volatile Matter (VM)	ASTM E872
Ash Content (AC)	ASTM D1102
Fixed Carbon (FC = 100-VM-AC-MC)	By difference

2.3 Ultimate Analysis

The ultimate analysis of PS, LDPE and PET respectively were carried out to determine their chemical composition. This was achieved using the Rutherford Back Scattering (RBS) technique in a Tandem Accelerator. The hydrogen content in each sample was however determined from the proximate analysis using the procedure employed by Adedaja *et al.* (2018) as shown in Equation 1.

$$H = 0.052 FC + 0.062 VM \quad (1)$$

2.4 Calorific Value

The higher heating value (calorific value) for each of the samples was determined in a bomb calorimeter (XRY-1C) using ISO 1928 standard.

2.5 Thermal Degradation

Thermogravimetric investigation was done on each of the samples by employing Perkins-Elmer TGA 4000 thermogravimetric analyzer. Approximately 10 mg of each of the plastic samples were introduced inside the TGA under a nitrogen gas flow rate of 100 mL/min. The sample was subjected to a heating regime from ambient temperature to 800°C using a heating rate of 20°C/min. The residual weight and the derivative weight of each waste plastic sample were then recorded as a function of temperature and time.

2.6 Kinetic Analysis

The kinetic parameters (activation energy and pre-exponential) factor of the pyrolysis of waste plastics (PS, LDPE and PET) were derived by employing the integration approach with the assumption that decomposition reaction via pyrolysis was a first-order reaction (Zhou *et al.*, 2009; Cepeliogullar and Putun, 2013; Oyedun *et al.*, 2015). The rate of decomposition of solid fuel is given by Equation 2, where x is taken as the conversion factor of the waste plastic which is calculated by Equation 3 with W_o as the initial weight of the sample, W_t as the weight of the sample at time t and W_f as the final weight of the sample. The rate constant of a reaction, k , can be expressed by the Arrhenius Equation 4. Equation 2 can be re-arranged further to give Equation 5, where A represents the pre-exponential factor with E representing the activation energy. Assuming that the rate of heating during the reaction, that is, $H = \frac{dT}{dt}$ is constant during the pyrolysis reaction, Equation 5 can be integrated and rearranged employing the Coats and Redfern integration method to give Equation 6. From Equation 6, a straight line graph is obtained when the left side of the equation was plotted against $1/T$ and the activation energy determined from the slope $-E/R$.

$$\frac{dx}{dt} = kf(x) \quad (2)$$

$$x = \frac{W_0 - W_t}{W_0 - W_f} \quad (3)$$

$$k = A \exp\left(-\frac{E}{RT}\right) \quad (4)$$

$$\frac{dx}{dt} = A \exp\left(-\frac{E}{RT}\right) (1 - x) \quad (5)$$

$$\ln\left[\frac{-\ln(1-x)}{T^2}\right] = \ln\left[\frac{AR}{HE}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (6)$$

3. RESULTS AND DISCUSSION

3.1 Proximate and Ultimate Analyses

The result for the proximate and ultimate analyses for each of the waste plastic is presented in Table 2. The moisture content of all waste plastic samples was very low with LDPE having a moisture content of 1.2 %, PS with a moisture content of 0.79 % while PET had a moisture content of 0.8%. This low percentage of moisture in waste plastics is due to the fact that waste plastics are already processed materials with moisture completely extracted from them and have no pore space for additional moisture to be introduced into their structure before, during or after use by the consumers (Durogbitan, 2019). The volatile matter content for all waste plastic samples ranged between 98.31 – 93.82 % with PS and LDPE having comparable volatile matter. The volatile matter content of solid fuels is indicative of the duration it will take such fuel to burn out. The presence of high volatile matter and low moisture contents in plastic materials suggests that plastic materials will be an effective source of fuel (Uzoejinwa, 2018). From the results obtained, all wastes plastics have a low percentage of ash ranging from 0.1-0.5%. The low percentage of ash in the samples suggests that all waste plastics are suitable for energy generation because a high percentage of ash has a negative effect on the calorific value of fuel (Durogbitan, 2019). The fixed carbon content of solid fuel is the leftover remaining after the moisture content, volatile matter content and ash content have been extracted. All the plastics investigated in this study have a very low percentage of fixed carbon.

The ultimate analysis of a sample gives a specification of the proportion of the various elemental composition of the sample as presented in Table 2. All the plastic samples considered in the study consist predominantly of carbon and hydrogen with a small proportion of oxygen and minute amount of nitrogen and sulphur. This shows that plastic materials are viable sources of fuel since they contain a very high proportion of carbon and hydrogen in their structure (Uzoejinwa, 2018). Rate of release of oxides of nitrogen and sulphur are determined by the proportion of nitrogen and sulphur in their structure since these oxides are released via the combustion of the solid fuel. From Table 2, all plastic waste samples in this study have trace quantities of sulphur and nitrogen content which shows that waste plastics are clean and environmentally friendly source of fuel.

Table 2: Proximate and ultimate analysis of PS, PET and LDPE

Characteristics	Variables	PS	PET	LDPE
Proximate analysis wt (%)	Moisture content	0.79	0.80	1.20
	Fixed carbon	0.80	4.74	-
	Volatile	98.31	94.36	98.30
	Ash	0.10	0.10	0.50
Ultimate analysis wt (%)	Carbon	93.32	61.05	93.76
	Hydrogen	6.14	6.28	5.74
	Nitrogen	0.04	-	-
	Oxygen	0.50	32.53	0.50
	Sulphur	-	0.14	-

3.2 Calorific Value

The heating value is a very critical factor since it shows the inherent energy contained in a fuel sample. The heating value also known as the calorific value of a fuel is dependent on its elemental composition and proximate analysis. The calorific values of the plastic waste samples are shown in Figure 1. The calorific value for PS (41.1 MJ/kg), LDPE (40.6 MJ/kg) and PET (22.1 MJ/kg) were obtained. The calorific value of PS and LDPE were high as a result of the high percentage of carbon and low percentage of ash in the solid fuels (Durogbitan, 2019). Meanwhile, the calorific value of PET was lower compared to that of PS and LDPE due to the higher content of oxygen. PS and LDPE have calorific values of 43.0 MJ/kg and 42.5 MJ/kg respectively. These values are comparable with the calorific values of conventional fossil fuels like diesel and gasoline (Ahmad *et al.*, 2014). This result shows that waste plastics have the potential for energy generation.

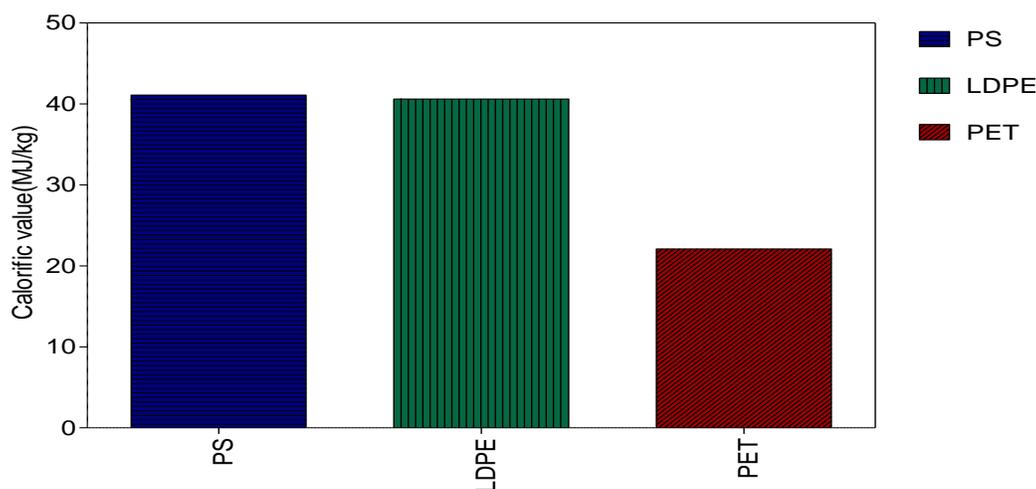


Figure 1: Calorific values of plastic wastes

3.3 Thermal Degradation Characterization of Post-Consumer Plastic

The thermogravimetric (TG) and differential thermogravimetric (DTG) profiles of the samples are illustrated in Figures 2 and 3 respectively. Profiles showing the percentage variation in loss of weight with reference to the initial weight of each sample are presented in Figure 2 while the rate of loss in weight is derived from the differential weight loss curve as presented in Figure 3. Generally, the thermal decomposition profiles for the plastic samples are very similar, however, temperatures at the beginning and end of thermal decomposition for each of the plastic samples are observed to be different indicating that each plastic sample exhibits different pyrolysis characteristics. Each plastic samples also show only one major weight loss stage as indicated by the single prominent peak shown in Figure 3. This is consistent with the behavior of plastic samples as corroborated by other researchers (Chattopadhyay *et al.*, 2008; Aboulkas *et al.*, 2009; Rotliwala and Parikh, 2011; Pan *et al.*, 2020). The weight loss of all samples is more than 97% at 800 °C, this is indicative of the fact that pyrolysis can be used to decrease drastically the volume of plastic waste in the environment.

Major reaction points as relating to the temperature of decomposition on TGA and DTG profile for each plastic sample showing the temperature interval of degradation (ΔT), peak degradation temperature (T_{max}) and mass loss rate (MLR_{max}) is presented in Table 3. It was observed that the decomposition of all plastic solid waste considered in the study occur at a temperature range of 368-546°C which is similar to the range specified for plastic materials (Na *et al.*, 2007). The maximum rate of loss in weight together with the peak temperature associated with it is also shown in Table 3. LDPE has the highest rate of change in weight of

30.98 wt%/min at a peak temperature of 519°C while PET has the least rate of mass loss of 24.23 wt%/min at a peak temperature of 462°C.

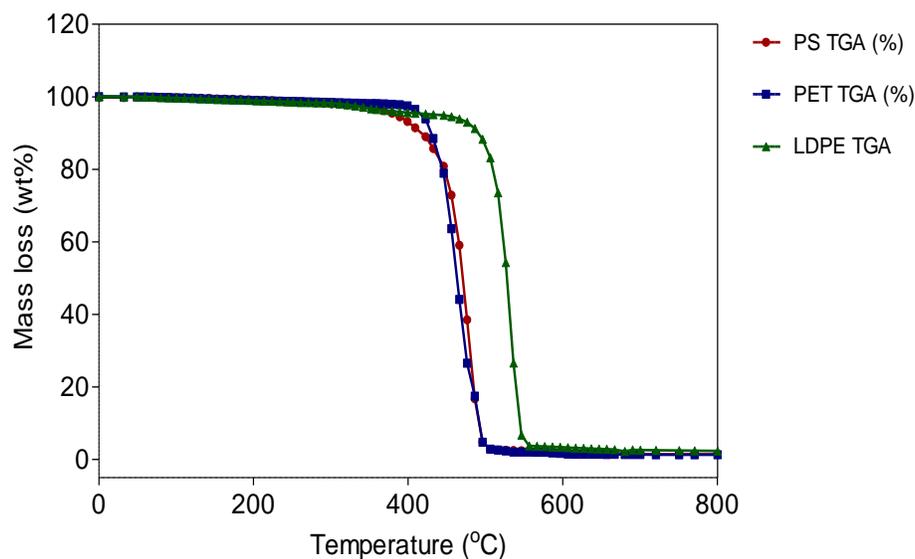


Figure 2: TG profiles of PS, PET and LDPE

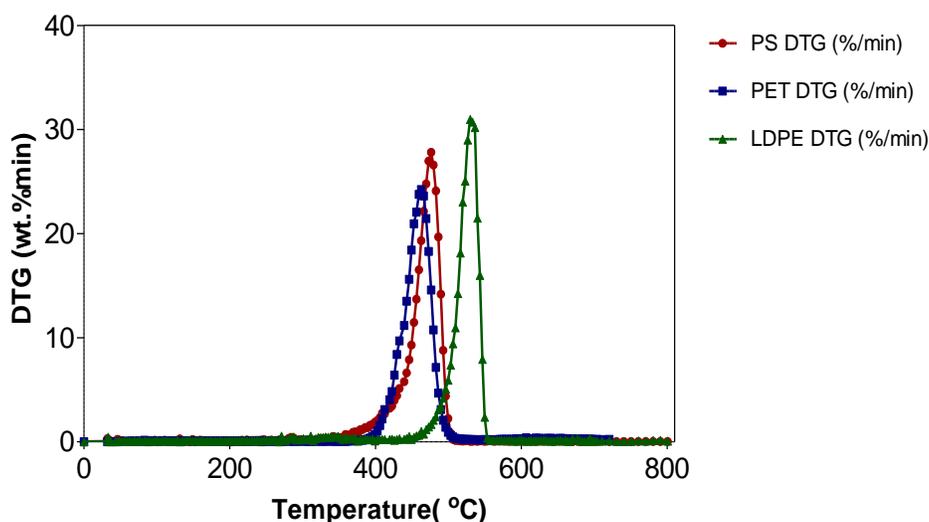


Figure 3: DTG Profiles of PS, PET and LDPE

Table 3: Degradation temperature range, peak temperature and maximum rate of mass loss of PS, PET and LDPE in nitrogen

Samples	$\Delta T(^{\circ}C)$	$T_{max}(^{\circ}C)$	$MLR_{max}(wt\%/min)$
PS	368-496	472	26.98
PET	399-496	462	24.23
LDPE	446—546	519	30.98

3.4 Pyrolysis Kinetic of PS, PET and LDPE

The linear regression models employed to determine the activation energy (E_A) of PS, PET and LDPE are presented in Figure 4. The R^2 values of all were above 0.9 indicating a good correlation coefficient of the linear model, therefore, the pyrolysis of the post-consumer plastics considered in the study can be

approximated as a first-order reaction. Similar observations were made by Wang *et al.* (2018) and Garba *et al.* (2017). The kinetic parameters of the samples are presented in Table 4. The apparent activation energy of LDPE (242.9 kJ/mol) was higher than that of PS (238.4 kJ/mol) and PET (189.05 kJ/mol) showing that LDPE requires more energy to break the inner bond in its structure compared with PS and PET. PET has the least activation energy which indicates that it is the least stable when subjected to a thermal regime (Espina *et al.*, 2015).

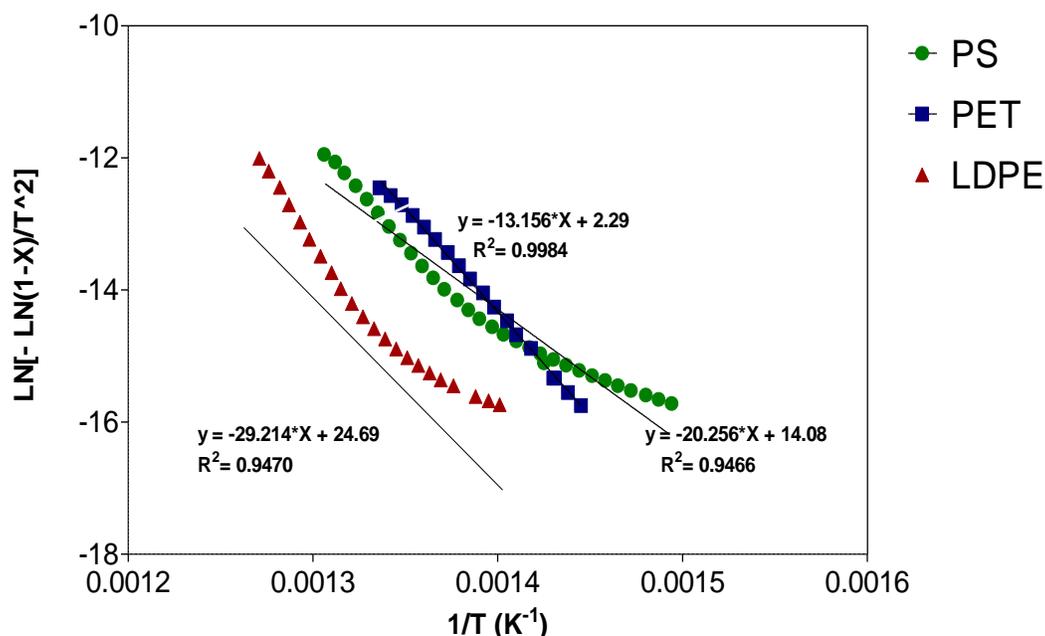


Figure 4: Pyrolysis kinetic curve for the degradation of LDPE, PET and PS

Table 4: Kinetic parameters of PS, PET and LDPE

Samples	E (kJ/mol)	R ²
PS	238.4	0.9466
PET	189.05	0.9984
LDPE	242.9	0.9470

4. CONCLUSION

This study carried out the decomposition behavior of some post-consumer plastic wastes to determine their pyrolysis kinetics. Prior to the decomposition study, the proximate and ultimate analysis revealed that all plastic wastes considered are suitable for conversion into fuel. Decomposition studies revealed that the degradation behavior of the plastic wastes occurred in a single phase. The total loss in weight of all samples considered was more than 97% at 800°C showing that pyrolysis technology can be employed to significantly reduce the volume of post-consumer plastic wastes. Observations also revealed that the experimental and calculated curves are in agreement showing that the single reaction mechanism model adopted is favorable.

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