



# **Production of Fuel Oil from Municipal Plastic Wastes Using Thermal and Catalytic Pyrolysis**

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## **Authors' contributions**

*This work was carried out in collaboration among all authors. Author DKO designed the study, performed the experiment, wrote the protocol and wrote the first draft of the manuscript. Authors OA and DFO managed the analyses of the study. Author OB managed the literature searches and wrote the final draft of the manuscript. All authors read and approved the final manuscript.*

## **Article Information**

DOI: 10.9734/JENRR/2020/v4i230120

### Editor(s):

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- Complete Peer review History: <http://www.sdiarticle4.com/review-history/54595>

**Original Research Article**

**Received 01 December 2019**

**Accepted 07 February 2020**

**Published 12 February 2020**

## **ABSTRACT**

Plastics have become an essential part of modern life today. The global production of plastics has gone up to 299 million tonnes in 2013, which has increased enormously in the present years. The utilization of plastics and its final disposal pose tremendous negative significant impacts on the environment. The present study aimed to investigate the thermal and catalytic pyrolysis for the production of fuel oil from the polyethylene plastic wastes. The samples collection for both plastic wastes and clay catalyst, sample preparation and pyrolysis experiment for oil production was done in Laroo Division, Gulu Municipality, Northern Uganda Region, Uganda. Catalysts used in the experiment were acid-activated clay mineral and aluminium chlorides on activated carbon. The clay mineral was activated by refluxing it with 6M Sulphuric acid for 3 hours. The experiment was conducted in three different phases: The first phase of the experiment was done without a catalyst (purely thermal pyrolysis). The second phase involves the use of acid-activated clay mineral. The

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third phase was done using aluminium chlorides on activated carbon. Both phases were done at different heating rates. In purely thermal pyrolysis, 88 mL of oil was obtained at a maximum temperature of 39°C and heating rates of 12.55°C /minute and reaction time of 4 hours. Acid activated clay mineral yielded 100 mL of oil with the heating rates of 12.55°C/minute and reaction time of 3 hours 30 minutes. While aluminium chlorides on activated carbon produced 105 mL of oil at a maximum temperature of 400°C and heating rates of 15.5°C /minute and reaction time of 3 hours 10 minutes. From the experimental results, catalytic pyrolysis is more efficient than purely thermal pyrolysis and homogenous catalysis (aluminium chlorides) shows a better result than solid acid catalyst (activated clay minerals) hence saving the energy needed for pyrolysis and making the process more economically feasible.

*Keywords: Activation; catalytic pyrolysis; fuel oil; hydrocarbon fuel; municipal wastes; plastics wastes; polyethylene; thermal pyrolysis.*

## 1. INTRODUCTION

Ineluctably in the modern world, today plastics have interwoven our life and life without plastics is exceptionally hard [1]. The global production of plastics increased by over 4% in 2012 and reached about 299 million tons in 2013 [2]. Plastic wastes are the third major component of municipal wastes in East African cities after organic waste and paper wastes. It was reported that in the year 2011, the plastic waste levels in East Africa was approaching 10% net weight of the total solid waste flows in major urban centres [3,4]. Presently, Landfill and incineration methods of plastic wastes disposal are being practiced within Africa and the western world through Municipal Solid Wastes (MSW) stream [5]. However, both methods are discerned to be posing negative impacts on the environment and are ultimately unsustainable [6]. Predominately, landfill is the method for disposing of plastic solid wastes (PSW) in the recent years [7]. Understanding this problem led to the development of several methods including chemical depolymerization, gasification, pyrolysis and catalytic degradation [8-11]. The products from tertiary recycling are often hugely varied depending on the process parameters utilized in plastic production [12-14]. Intrinsically, vast number of researches have been performed to ascertain the conditions which produce the highest value of product yields.

Ahmad, et al. [15] explored the pyrolysis study of High-density polyethylene (HDPE) using micro steel reactor. The pyrolysis temperatures were within 300–400°C at a heating rate of 5–10°C/min. Nitrogen gas was used as the fluidizing medium. From their experiment, the authors found that the highest total conversion happened to be at 350°C with liquid as the dominant product yield (80.88 wt.%). The solid

residue was very high at 300°C (33.05 wt.%) but its amount reduced to 0.54 wt.% at the highest temperature of 400°C. On the other hand, Kumar & Singh [16] conducted a thermal pyrolysis study of HDPE using a semi-batch reactor at a higher temperature range of 400–550°C. It was observed that the highest liquid yield (7.08 wt.%) and gaseous product (24.75 wt%) occurred at a temperature of 55°C while wax started to dominate in product reaction at the higher temperature of 500–550°C. The dark brownish oil obtained from the pyrolysis had no visible residue and the boiling point was from 82 to 352°C. The authors further stressed that the mixture of different oil component such as gasoline, kerosene and diesel in the dark brownish oil matched the properties of conventional fuel. Besides, its sulfur content was very low (0.019%) making HDPE pyrolytic oil a cleaner fuel which is environment-friendly.

As an innovative way to recover energy and reduce waste, pyrolysis of Low-Density Polyethylene (LDPE) to oil production has received much attention by vast researchers in recent years [17–22]. For instance, Bagri & Williams [23] investigated the LDPE pyrolysis in the fixed-bed reactor at 500°C with a heating rate of 10°C/min. The experiment was done for 20 min and nitrogen was used as fluidizing gas. It was observed that high liquid yield of 95 wt% was obtained with low gas yield and negligible char. In the present study, the investigation on catalytic pyrolysis of a mixture of LDPE and HDPE plastic wastes was conducted to determine the potentiality of the locally available clay catalyst for plastic wastes degradation. The degradation of plastic wastes with catalytic pyrolysis is very important as it is the most economically viable and sustainable process of producing fuel oil [12–14]. Therefore, the present study focused on the investigation of activated

montmorillonite clay in catalyzing the pyrolysis reaction of the blend of LDPE and HDPE plastic wastes and comparing it with the conventional catalyst (Aluminium chloride).

## 2. MATERIALS AND METHODS

### 2.1 Sample Collection

The waste plastics used in this experiment consisted of a mixture of 100 g HDPE and 100 g LDPE resins which were collected from within Laroo division rubbish pit around Gulu University main campus. The samples were manually collected from the designated area, packed in sacks and carried to the chemistry laboratory. From the laboratory, the waste plastics were sorted according to their resin type as indicated by the resin identification codes and placed in different boxes. The dirty plastic wastes were washed with water and detergent solution, dried and then kept for further processing.

### 2.2 Pelletization of Plastic Wastes

This is a process of forming plastic pellets from the light and bulky plastic materials. This helps reduce their surface area and makes it easy to be transported and packed inside the reactors. During this process, the cleaned plastics were cut into small pieces and placed on the hot plate heating machine covered by aluminium foil, and heated to a temperature of about 150°C and rolled with a piece of metal into a ball like structure (Fig. 1). The pellets were then cooled and stored for the pyrolysis experiment.



**Fig. 1. Pellets of a mixture of HDPE and LDPE plastic wastes**

### 2.3 Preparation of the Natural Catalyst

The conventional catalyst material used for comparison was a Lewis acid-base catalyst (Aluminum chlorides) supported by activated carbon both obtained from the local market.

While the raw montmorillonite clay (Fig. 2) is locally available and was collected from within Gulu municipal swamp valley beds.



**Fig. 2. Raw montmorillonite clay**

The raw montmorillonite clay material obtained was massed up using a wooden roller and cut into small cubes (approximately 1 cm<sup>3</sup>). The cubes were heated in the universal hot air oven at 150°C for six hours, allowed to cool for one day and crushed into a fine powder using a mortar and pestle. The mixture was sieved to remove the gravels and small stones and the fine powder mixed with 6 M sulfuric acid in a glass beaker, stirred and the mixture refluxed for 3 hours and vacuum filtered. The residue in the filter paper was washed thoroughly using distilled water several times, and the clean residue was tested for the presence of sulfates using acidified barium nitrates solutions to ensure that no sulfate ion is present in the activated clay sample. It was placed inside the universal hot air oven for another day for drying. The activated montmorillonite clay produced (Fig. 3) was then made into a paste by blending it with starch powder and distilled water which was then rolled into small cubes, dried and kept for the experiment. The advantage of pelletizing the catalyst powder was to reduce the pressure drop in the catalytic bed. While the blending of the activated clay with a starch powder was to increase the void fraction between the inter-particles of the catalyst pellets



**Fig. 3. Acid activated montmorillonite clay**

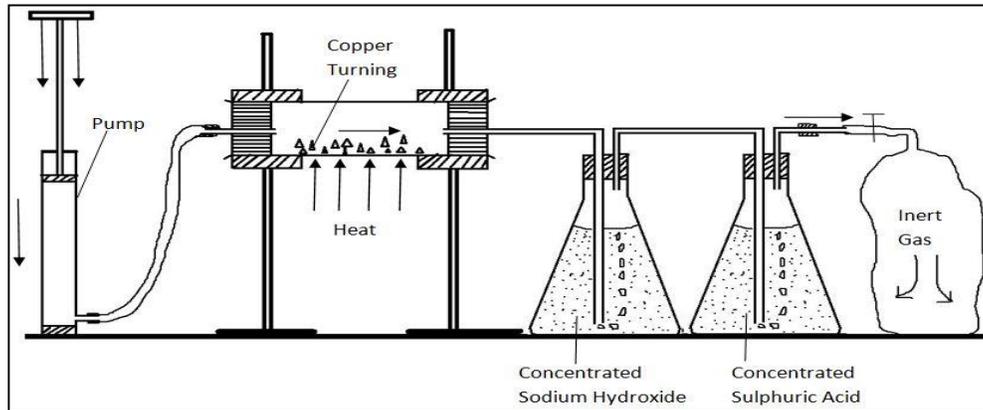
### 2.4 Laboratory Preparation of Inert Gas

The inert gas was prepared in the laboratory by removing all the reactive gases: oxygen, carbon dioxide and water vapor from the atmospheric air by using simple laboratory technique as illustrated in Fig. 4. The air was pumped using a bicycle pump into the combustion chamber containing copper turning which removed oxygen from the air. Then, it was passed into a conical flask containing concentrated sodium hydroxide which eliminated carbon dioxide from the air. Lastly, it was passed into a conical flask containing concentrated Sulphuric acid to remove water vapor and the dry inert gas was collected and stored for fuel oil production.

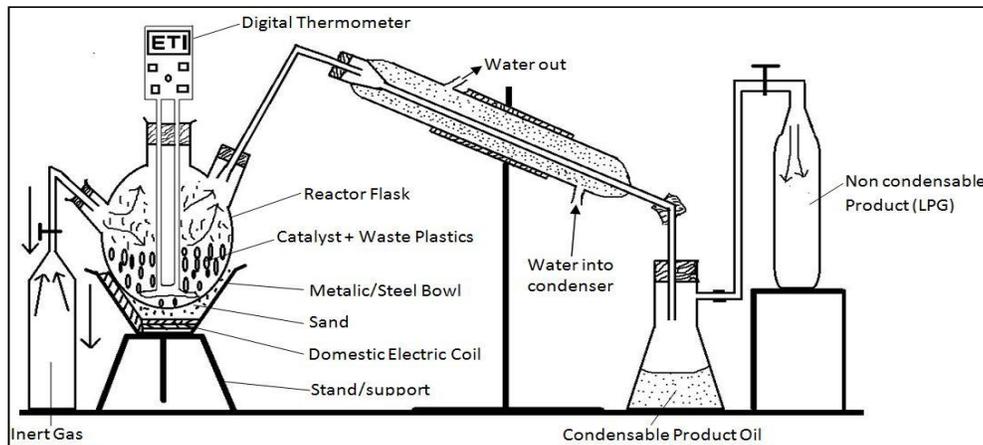
### 2.5 Experimental Setup for Fuel Oil Production

A mixture of HDPE and LDPE plastic resins (1:1 w/w, 200 g) were fed into the reactor flask and 20

g of acid-activated clay catalyst (10:1 w/w) was added. The electrode of the thermocouple thermometer was inserted in the reactor flask and the tip was immersed in the fed stoke catalyst mixture from the vertical opening and sealed completely as shown in Fig. 5. The reaction flask was placed on sand contained in a steel basin and was heated electrically. The inert gas was allowed to pass through the set up for some time to drive out air and replacing it with the inert nitrogen gas. After flashing the system, the Teflon bag was connected to the sidearm of the conical flask. When all the connection is made, the system becomes isolated from the surrounding in terms of air movement. The power was switched on, the temperature and time taken for the fed stoke to melt were recorded. The temperature and amount of oil produced were recorded at an interval of 3 hours. The dark brownish oil produced (Fig. 6) was measured using a measuring cylinder.



**Fig. 4. Set up for laboratory preparation of inert gas**



**Fig. 5. Experimental setup for fuel oil production from plastic wastes**



**Fig. 6. Dark brownish fuel oil produced from plastic wastes**

Using the same experimental setup in Fig. 5, another two sets of experiments were performed by repeating the fuel oil production experiment with aluminium chloride on activated carbon as catalyst instead and without catalyst (purely thermal pyrolysis).

## 2.6 Data Analysis Method

The data collected were fed in excel and statistically analyzed to determine the correlation between temperature and the amount of oil collected for the three pyrolysis conditions.

## 3. RESULTS AND DISCUSSION

The pyrolysis of plastic wastes was investigated as an effective approach to recycle municipal plastic wastes. Plastic wastes can easily be converted into fuel oil using locally available materials.

Table 1 indicates the yields attained after cracking a mixture of HDPE and LDPE at varying temperatures. The degradation of plastic wastes started at temperatures above 150°C for all the three conditions investigated as shown in Fig. 7. It was observed that for purely thermal pyrolysis, the degradation started at the temperature of 205°C and a slight increase in temperature from 205°C to 222°C at an average heating rate of 12.5°C /minute produced 5 mL of oil. However, for catalytic pyrolysis, the drop of oil started at much lower temperatures (170°C) and (185°C) using aluminium chlorides catalyst and acid-activated clay minerals respectively. An increase in temperature from 170°C to 200°C produced 15 mL of oil which is 7.5% conversion with aluminium chloride catalyst. While 10 mL of oil was collected that is 5% conversion for the

temperature increase from 185°C to 200°C when using acid-activated clay catalyst. These results obtained were lower than those reported by Motevasel, et al. [12] who found out that thermal degradation of a mixture of plastic wastes starts at 370°C with less than 10% conversion but with up to 40% conversion when using a catalyst. The big difference is due to the variation in temperature distribution in the reactor, hence, the plastics in contact with the reactor absorbs heat faster and start degrading. At some points, even when melting and volatilization had already started, the thermo-couple would still be indicating low temperatures. Nevertheless, it was possible to determine that for thermal pyrolysis, the degradation temperature is higher compared to those of catalytic degradations. This is basically because the catalysts offer alternative pathways of lower activation energies. The result also indicates that catalytic degradations registered a higher increase in the amount of oil collected for the corresponding increase in temperature than purely pyrolysis process.

From Fig. 8, it can be seen that for every temperature increase attained, the quantity of oil is greater for the catalytic conversion when compared to the purely thermal conversion process. This is in accordance to the results obtained by Motevasel, et al. [12] who found that oil yield increases with temperature when using the catalyst. However, at very higher temperatures (350–400°C), even thermal degradation becomes almost as efficient as catalytic degradation. Therefore, it can be concluded that catalytic degradation is an efficient method of the pyrolytic conversion of plastic wastes into fuel. Notwithstanding, catalytic conversion using aluminium chloride (homogenous catalyst) yielded slightly higher amounts of oil compared to using an activated catalyst (heterogeneous catalysis). This is because the heterogeneous catalyst having a solid particulate matter interferes with the heat conductivity in the reactor which affects the overall distribution of heat in the reactor and causes low degradation at a particular temperature.

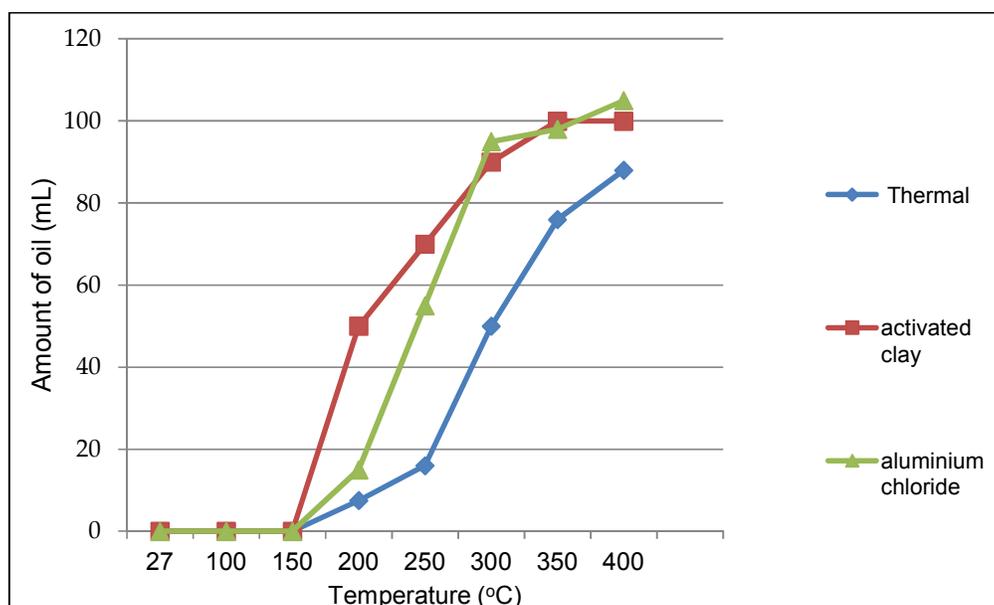
The aluminium chlorides catalyst works in the homogenous state, whereby on adding the catalyst in the reacting vessel, it gets dissolved in the molten plastics and becomes in a uniform phase with the reacting mixture without interrupting the process of heat conductivity. Therefore, heat conductivity and distribution are faster when compared to a solid acid

(heterogeneous catalyst). So, the product formed at a specific temperature will be higher compared to that of the solid acid catalyst in heterogeneous phase. However, the problem with using the homogenous catalyst in the pyrolysis reaction is its irrecoverability after the reaction completion,

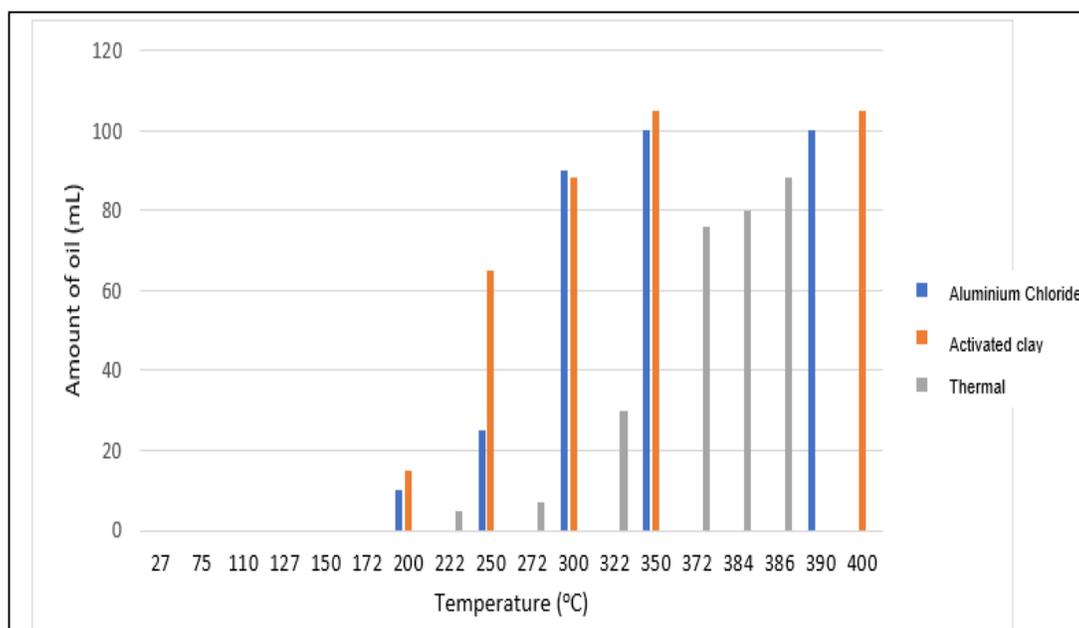
therefore, its disposal is unsafe to the environment. While solid acid such as activated clay catalyst is recoverable after the reaction and can be reused and its disposal does not cause any damage to the environment. It is a green chemical transformation.

**Table 1. Pyrolysis of plastic wastes to fuel oil**

Temperature (°C)	Amount of oil (mL)		
	Acid activated clay catalyst (Reaction heating rates = 10.5°C/min)	Aluminium chloride catalyst (Reaction heating rates = 15.5°C/min)	Thermal pyrolysis (Reaction heating rates = 12.5°C/minute)
27	0	0	0
75	-	0	-
110	-	0	-
127	0	-	0
150	0	0	-
172	-	-	0
200	10	15	-
222	-	-	5
250	25	65	-
272	-	-	7
300	90	88	-
322	-	-	30
350	100	105	-
372	-	-	76
384	-	-	80
386	-	-	88
390	100	-	-
400	-	105	-



**Fig. 7. Line graph showing the variation of the amount of oil collected with pyrolysis temperature**



**Fig. 8.** Bar graph showing the amount of oil collected at varying temperatures

#### 4. CONCLUSION

The pyrolysis of plastic wastes was investigated as an effective approach to recycle municipal plastic wastes. Plastic wastes can easily be converted into fuel oil using locally available materials. In the experiments conducted it was found that more oil product was obtained using catalytic pyrolysis at the specific temperature and the degradation of the plastics started at a lower temperature compared with purely thermal pyrolysis. Therefore, catalytic pyrolysis is a better method of conversion of plastic wastes to fuel oil. Acid activated clay mineral is a good heterogeneous cracking catalyst since it is recoverable after use and its final disposal poses no threats to the environment. Moreover, it has good catalytic activity compared to a homogenous catalyst which is unrecoverable after use and the disposal of the residue poses a significant negative impact on the environment. The degradation of plastics is favored at high temperature of 340-400°C, this is the temperature ranges where more liquid oil products are formed. Profoundly, further study should be directed toward the analysis of the chemical composition of the fuel oil produced. additionally, further study should compare the quantity and quality of oil produced with acid-activated clay catalyst and with catalytic pyrolysis involving metal oxide and zeolites.

#### ACKNOWLEDGEMENTS

The Authors (DKO), (OA) and (DFO) acknowledge the management of Gulu University for the administrative and technical support. Author (OB) acknowledge ACE II-PTRE of Moi University for the administrative support.

#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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