Article

Effects of Process Factors on the Pyrolysis of Waste Sac Bags over Corn Cob Activated Carbon Catalyst into Aviation Range Hydrocarbons

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Abstract

In the present study, design of experiment approach, specifically the central composite design (CCD) in Design Expert 12.0 was used to obtain a design matrix for the pyrolysis of waste sac bags over corn cobs activated carbon catalyst. Statistical method was used to analyze the effect of temperature and catalyst to feed ratio on the yield of fuel obtained. Analysis of variance of the results obtained show that both parameters are significant in the process. It was also observed that the optimal yield of fuel oil was 64.54 % and this occurred at a temperature of 600°C and a catalyst to feed ratio of 0.1. The specific gravity of fuel oil obtained from the pyrolysis of waste sac bags with the local catalyst, fuel obtained without catalyst, commercial cooking kerosene and commercial aviation kerosene were 0.771, 0.794, 0.806, and 0.779 respectively. The specific gravity of the fuel oil obtained with the local catalyst is very close to that of commercial aviation kerosene, this means that it can be used aviation fuel. Results obtained from gas chromatography of the samples further confirms that, the fuel oil obtained from the use of corn cobs activated carbon catalyst contains hydrocarbons of the aviation fuel range. This study shows that a circular economy is feasible, and that value can be obtained from waste plastics if the proper process and process factors are utilized.

Keywords: Waste plastics; Agro waste; Aviation range hydrocarbons; Pyrolysis.

1. Introduction

Plastics are inexpensive and long-lasting; nonetheless, reducing waste plastic and increasing waste plastic recycling knowledge are vital, if we are to address the problem of waste plastic pollution in our world today. Statistics show that approximately 8 million tonnes of waste plastics end up in the ocean annually, this figure could double if nothing is done about it by the year 2025, and by the year 2050 the waste plastics in our oceans would be more than the fishes ^[1]. This is how much waste plastics impacts on the environment. Some researchers have been able to develop bio-plastics ^[2], this sounds good since they are from plants- based sources such as flaxseed, however a United Nations report in 2015 states that once the polymer is created, the material properties are the same, which implies that the resulting material is no better that any other form of plastic, because bio-plastics can't be recycled nor composted. Waste sac bags are common in the agricultural sector, they are used in bagging agricultural products ready to be transported, and also for storage purposes. Nigeria is a country known for agriculture, this has made these bags very common, because agricultural products like rice, beans, wheat, flour, are packaged in sac bags. Sac bags shred with time, and once they are shredded, they can no longer be used for storage, and since they are not biodegradable, disposing them becomes a problem. Waste plastics have been managed in a variety of ways over the years, including burning, landfill disposal, and recycling into valuable items. These strategies are insufficient for establishing and maintaining a circular economy for plastics ^[3]. Because plastic garbage is bulky and combustion releases hazardous compounds into the atmosphere, disposal of plastic waste in landfills and by incineration is costly and pollutes the environment ^[4]. In contrast, depending on the plastic type, ecology, ease of separation from other waste materials, and processing cost, values can be recovered from waste plastics in the form of materials recycling or energy ^[5]. Ref ^[6] obtained diesel range products from Polyethylene, polypropylene and polystyrene waste plastics. Ref ^[7] obtained gasoline range fuels from the pyrolysis by using polyethylene and polypropylene as raw materials ^[8], converted used tyres to liquid fuels ^[9-10], used discarded sachet water bags to obtain fuel oil, but the conversion of sac bags into fuels has not been fully researched.

Aviation range hydrocarbon is a middle distillate of crude oil fractional distillation, specifically kerosene range ^[11]. Generally, it consists of carbon chains ranging from C₈-C₁₆, which also varies depending on the Jet fuel type ^[12]. Soják *et al.* used GC-MS to characterize the products of thermal cracking of polyethylene and polypropylene ^[11]. Aviation fuels are a multicomponent mixture of paraffins, naphthenes or cycloparaffins, aromatics and olefins compounds. The major components of aviation fuel are paraffins, isoparaffins and cycloparaffins, which constitute about 70-85 % of the fuel. These major components are responsible for reducing the freezing point of jet fuel, the aromatic component of the fuel is responsible for improving the energy density, shrinkage of aged elastomer seals and also aid in reducing fuel leakage problems ^[13].

Recent trends in sustainable energy research show that researchers have recorded success in the conversion of waste plastics into hydrocarbon fuels via the process of pyrolysis. During the pyrolysis of waste plastics, long chain hydrocarbons are thermally broken down into short chain hydrocarbons in the absence of oxygen, into liquids, gases and char, this process is called thermal pyrolysis. Introducing a catalyst in the process of pyrolysis can lower the temperature, reduce the reaction time and also, selectively increase the yield of specific products, which can be used as fuel oils and petrochemical feedstock ^[14]. This type of pyrolysis is called catalytic pyrolysis. The type of fuel obtained largely depends on the production route or pathway. Ref ^[15], explained the various pathways for the production of aviation fuel, which include: thermochemical process, biomass to liquid process, Fischer Tropsch process, hydro-processing, biochemical processes, and bio alcohol production. Pyrolysis is a thermochemical process that could be classified as thermal or catalytic pyrolysis. Thermal pyrolysis uses high temperature to decompose waste plastics into liquids, gases and char while catalytic pyrolysis involves the use of a catalyst to aid the thermal process. Ref ^[16], stated that thermal pyrolysis gives a wide range of products and requires high temperatures and that introducing a catalyst into the process helps in the selectivity of the products. A lot of work has been done to convert the waste plastics into useful fuels thermally ^[17-18], by using a catalyst ^[11,19-20].

Catalysts play a very major role in chemical processes; they help to increase or decrease the activity of chemical reactions. Literature has shown that a good number of catalysts such as zeolites, nano-crystalline zeolite (HUSY, n-HZSM-5 HMOR), silica-alumina, FCC, MCM-41, Kaolin, cement powder and others have been used in pyrolysis process to obtain hydrocarbon fuels from waste plastics ^[18]. These catalysts vary in their properties and also affect the product distribution and reaction rates differently. A recent study by ^[19], suggested that activated carbons are good catalysts for selectively obtaining Jet fuels from waste polyethylene pyrolysis, this is due to their porous structure, and these pores trap oils which promotes further cracking. In this paper, activated carbons obtained from corn cobs is used as catalysts for the production of aviation fuel range hydrocarbons from waste sac bags. The advantage of these carbonaceous materials is that they are readily available in the environment, easy to prepare and cheaper than crude oil processing into petrochemicals in the form of plastics ^[20].

Different parameters impact on the pyrolysis of waste plastics, these parameters are temperature, residence time, catalyst type, catalyst to feed ratio, heating rates, and inert gas ^[21]. This work provides the analysis of temperature and catalyst to feed ratio in the pyrolysis of waste sac bags over corncobs catalyst into aviation range hydrocarbon fuel.

2. Materials and methods

50 kg sac bags were obtained from rice traders in Oroigwe, Port Harcourt. About 100 kg fresh corn was collected from the farm in Etchie, Port Harcourt, Rivers State, Nigeria. Phosphoric acid 85% pure, distilled water and the acids were applied in the preparation of the catalyst. Commercial cooking kerosene and commercial aviation kerosene were obtained for the purpose of comparison and test analysis.

2.1. Preparation of activated carbon from corn cobs

To get the range of hydrocarbons targeted, it is beneficial to use a catalyst. Freshly cut corns from the farm were peeled and deseeded manually with hand, the cobs were dried at a temperature of 110°C, for four hours to reduce the moisture content before chemical activation. The dried corn cobs were cut into smaller sizes of about 5 cm using a kitchen knife in order to increase the surface area and aid the drying process. 200 g of the corn cob was soaked in a solution of phosphoric acid overnight (for about 12 hours). The corn cobs were removed from the acid solution and washed with water before drying them at an activation temperature of 600°C for one hour. The dried samples are then grinded with an electric bender (Original Millennium Nakai Blender Model 333 special) to reduce its particle size. An electronic sieve shaker (Cisa BA 200N, 01996, Spain) was used to determine the particle sizes. The activated carbon sample was stored in an airtight container.



Fig 1. Corn cobs before activation Fig 2. Corn cobs after activation Fig 3. Activated carbon in powder form

2.2. Production of fuel oil via pyrolysis

50 g of waste polypropylene plastic and corncobs catalysts (5 g or 15 g) were fed into a stainless-steel batch reactor, the reactor has two lids, one at the top connected to a pipe that collects vapor from the reactor. Another at the side through which the nitrogen gas gets into the reactor. The reactor was tightly closed and put into the muffle furnace (JP Selecta, 582543 S/N, 230 VAC, 00-C/2000367, Spain), and the furnace door was closed. The muffle furnace has a PID temperature regulator. The pipe connected to the top lid terminates into a reflux condenser where the vapor is condensed. The condensate is collected in an oil collector having an opening by the side through which the uncondensed gases exit. The yield of fuel oil obtained was calculated by weighing the fuel oil collected in a weighing balance, and the percentage yield calculated using equation (2). The oil was stored in a test bottle.

Mass of waste plastic + mass of catalyst = mass of fuel oil +mass of gas +mass of char (1)

Percentage yield of fuel oil =
$$\frac{\text{mass of uer on}}{\text{mass of waste plastic+mass of catalyst}} \times 100$$
 (2)



Fig 4. Experiment set up. 1. Nitrogen gas bottle 2. Furnace 3. Stainless steel reactor 4. Vapour outlet 5. Reflux condenser 6. Tap water 7. Cooling water outlet 8. Ice bath 9 Oil-collector 10. Uncondensed gas outlet 11. Tap water sink outlet



Fig 5. Samples of fuel oil obtained

2.3. Analysis of fuel oil from activated carbon catalyzed pyrolysis of waste polypropylene

To investigate the impact of temperature and catalyst to feed ratio on the yield of fuel oil obtained, the two factors were varied – temperature at two levels (600°C, 640°C), and catalyst to feed ratio also at two levels (0.1, 0.3) using the CCD design in design expert 12.0. The response variable is the yield of fuel oil at set temperature. The liquid fuel oil obtained from the catalytic pyrolysis was characterized: specific gravity and density were obtained using Metler Toledo Densito 30PX (ASTM D1298). Agilent 7890B GC was used to identify the compositions and distribution of carbons in the fuel oil obtained.

3. Results and discussion

3.1. Experimental design and yield of fuel oil from catalytic pyrolysis of waste sac bags over activated corn cobs catalyst

Table 1 represents the yield of fuel oil are set conditions, the table shows that a maximum yield of 64.54 % fuel oil was obtained at a temperature of 600°C and catalyst to feed ratio of 0.1 while a minimum yield of 18 % was obtained at a temperature of 640°C and catalyst to feed ratio of 0.3. The P-value from the Table 2 shows that temperature and catalyst to feed ratio are very significant parameters to be considered in the pyrolysis of waste polypropylene using activated corn cobs catalysts. Estimates of coefficient are shown in Table 3, the coefficient estimates were used to generate the normalized equation model (Equation 3), and this equation can be used to determine the yield of fuel oil at any set temperature for this catalytic pyrolysis process.

Yield of fuel oil at set temperature = $8.16A^2 - 16.13A - 5.59B + 30.37$ (3)

Std	Run	Temperature (°C)	Cat : Feed	Yield of fuel oil (wt %)
2	1	640	0.1	28.55
1	2	600	0.1	64.54
7	3	620	0.1	34.73
6	4	640	0.2	21.78
8	5	620	0.3	26.78
5	6	600	0.2	51.03
3	7	600	0.3	41.53
4	8	640	0.3	18.00
9	9	600	-	16.6

Table 1. Yield of fuel oil at set temperature

Table 2. Anova for reduced quadratic model for yield of fuel oil

Source	SS	D.f	Mean	F-value	P-value
Model	1847.76	3	615.92	64.41	0.0008
Α	1560.74	1	1560.74	163.23	0.0002
В	186.27	1	187.27	19.58	0.0115
A ²	99.76	1	99.76	10.43	0.0320
Residual	36.25	4	9.56		
Cor. total	1885.01	7			

A - Temperature; B - Catalyst to feed ratio; SS- sum of squares

Factor	Coe. estimate	D. f	S.E	CI low	Cl high
Intercept	30.75	1	2.19	24.68	36.82
Α	-16.13	1	1.26	-19.63	-12.62
В	-5.59	1	1.26	-9.09	-2.62
A ²	8.16	1	2.52	1.15	15.16

Table 3. Coefficient of estimates

Table 4 compares the predicted values for yield of fuel oil and the actual yield of fuel oil. It shows that, predicted R² of 0.9250 is very close to the adjusted value R² of 0.9645, the difference is 0.03 which is less than 0.2, this confirms that model reduction or modification is not required and that the model terms used are significant. The value of 19.8626 for adequate precision is good, this implies that the model can be used to explain the design space, Design expert 12.0 recommends an adequate precision value that is greater than 4. Adequate precision simply is a measure of signal to noise ratio, (Design expert 12.0 Tutorial). The goodness of fit shows that the predicted values are very close to the actual values obtained for the yield of fuel oil.

Table 4. Goodness of fit

Std Dev	Mean	CV%	R ²	Adj. R ²	Pred. R ²	Ad. precision
3.09	36.87	8.39	0.9797	0.9645	0.9250	19.8626

3.2. Effect of temperature on the yield of fuel obtained at set temperature

Figure 6, shows the impact of temperature on the yield of fuel oil obtained at set temperature, the maximum yield of the fuel oil of 64.54 % was obtained at a temperature of 600°C, it was observed that the yield consequently reduced as the temperature was increased from 600°C to 640°C. A maximum yield of 79.85 % of liquid fuel was obtained at a temperature of 450°C ^[22], this disparity in the results obtained could be as a result of the different types of reactors used and also the different types of catalysts used.



Fig 6. Effect of temperature on the yield of fuel oil

3.3. Effect of catalyst to feed ratio on the yield of fuel oil obtained at set temperature

In order to investigate the impact of catalyst to feed ratio on the yield of fuel oil obtained, the catalyst to feed ratio was varied at 0.1, 0.2 and 0.3. Figure 7 shows that the least catalyst to feed ratio of 0.1 resulted in the maximum yield of liquid fuel of 64.54 %, this is in agreement with the study by ^[23], in which FCC catalyst was used a decreasing yield of condensate fuel was observed as the catalyst to feed ratio was increased from 0.1 to 0.6. This indicates that intense cracking of the waste plastic took place at a catalyst to feed ratio of 0.1. When waste polypropylene plastics and was pyrolyzed with commercial activated carbon in a stainless steel reactor operating at a temperature range of 100 - 400, 34.40 % fuel oil containing aviation

range hydrocarbons was obtained in 6.5hrs, ^[24]. The yield obtained is less than the value of 64. 54 % of fuel oil obtained in this work, this shows that the operating conditions used helped optimize the process. 40 % of fuel oil was obtained with polypropylene waste plastics in a thermal batch within a residence time of 75 minute, this value is lower than the value of fuel oil obtained in ^[23], and the present study, because no catalyst was used. In a study to obtain Jet fuel range hydrocarbons, corn stover activated carbon was used in the pyrolysis of waste polyethylene waste plastics a yield of 66.5 %-73.1 % of Jet fuel range hydrocarbons was obtained in facile tube fixed bed reactor at a residence time of 6.5 hours ^[19]. The yield of fuel oil obtained is slightly higher than the value obtained in this work. For this work a residence time of 1.5hours was used to achieve pyrolysis.

Figure 8 is a three dimensional plot, showing the effect of temperature and catalyst to feed ratio on the yield of fuel oil. This plot clearly shows the point that the optimum yield of fuel oil was obtain.



Fig 7. Effect of catalyst to feed ratio on the yield of fuel oil



Fig 8. Effect of temperature and catalyst to feed ratio on the yield of fuel oil

3.4. Comparison of pyrolysis oil produced with commercial kerosene and aviation fuels

To compare the fuel oil samples obtained in this work with commercial cooking kerosene and commercial aviation kerosene, it was necessary to determine the distribution of carbons in the fuel oil sample obtained from the experiments, GC analysis was done on the sample A (fuel oil obtained from catalytic pyrolysis) and compared with the GC results obtained when the pyrolysis was done without a catalyst (sample B), with commercial cooking kerosene (sample C) and commercial aviation kerosene (sample D). From the result obtained, the fuel oil sample B contains C_8-C_{16} hydrocarbons and with further treatment can serve as an alternative for both cooking kerosene and aviation kerosene. The apparent molar mass for the

samples were also obtained to be 138.71 g/gmole, 144.90 g/gmole 145.25 g/gmole and 148.86 g/gmole for samples A, B, C, and D respectively. The density and specific density of the fuel samples were also determined, this was done to compare with that of commercial cooking kerosene and commercial aviation fuel. The result in table (5), shows that the fuel oil obtained from the catalytic pyrolysis of waste polypropylene plastics with corn cobs activated carbon is very close to the specific gravity commercial aviation kerosene than the specific gravity of commercial cooking kerosene.

Table 5. Density and specific gravit

Property	Unit	Sample A	Sample B	Sample C	Sample D
Density	kg/m³	0.768	0.792	0.805	0.778
Specific gravity		0.771	0.794	0.806	0.779



Fig 9. Carbon distribution in fuel oil obtained from the pyrolysis of waste sac bags using corncobs activated carbon (sample A)



Fig 10. Carbon distribution in fuel oil obtained from the pyrolysis of waste sac bags (sample B)



Fig 11. Carbon distribution in fuel oil of commercial cooking kerosene (sample C)



Fig 12. Carbon distribution in fuel oil of commercial aviation kerosene (sample D)

Fig 13 shows the various compositions of carbons found in each fuel sample, this clearly indicates that the fuel oil obtained from pyrolysis of waste polypropylene sac bags contain the different carbon ranges, but the composition of C_8 - C_{16} is very high. This implies that it contains the aviation range hydrocarbons. The fuel oil obtained in this study is still in its crude state and has not been refined or treated, unlike the commercial aviation kerosene which has been refined to a high purity kerosene.



Fig 13. Composition of hydrocarbons in fuel oils

4. Conclusions

Analysis of temperature and catalyst to feed ratio on the yield of fuel oil obtained when waste sac bags undergo pyrolysis under activated corn cobs catalysts shows that they both have an effect on it. The optimum yield of fuel oil of 64.54 % was obtained at a temperature of 600°C and catalyst to feed ratio of 0.1. Temperatures above 600°C produced less quantity of fuels. Increasing the catalyst to feed ratio above 0.1 also produced a lesser quantity of fuel, a model equation that can calculate the yield of fuel oil for the catalytic pyrolysis of was sac bags at any temperature and catalyst to feed ratio have been developed (Equation 3). Gas chromatography of fuel oil obtained shows that it is similar to aviation fuel and the peaks show that it has a high composition of C_8-C_{16} hydrocarbons.

Waste sac bags as one of the many plastic wastes is an environmental challenge could be used to produce aviation fuels. In this approach, environmental remediation could be enhanced by turning waste into new resources and materials, which has been experimentally demonstrated in this work. The first approach is the sustainability approach by recycling and reusing the waste sac bags (and possible for other kinds of plastic waste). The second approach is the circularity concept of reusing agro-waste in this case corn cob as catalyst additives or supports by providing biomass conversion process in adding value to conventional catalyst technologies.

Conflicts of interest

The authors declare that there is no conflict of interest.

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