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Original Research Article

Blend Property of the Extracted Diesel Component by Hydropyrolytic Depolymerization of LDPE, LLDPE, MDPE and HDPE type of Polyethylene Mix

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Abstract

With the fast depleting conventional energy reserves, the thrust for developing newer and alternative energy sources is the key research area worldwide. With the increase in the pollution level there is a need for searching a sustainably developed and environmental friendly energy source. The municipal solid waste has become one of the major pollutants worldwide. About twenty percent of the municipal waste comprises of the different types of polyethylene waste. The process of conversion of the polyethylene waste into useful fuel has become one of the emerging fields, countries are working worldwide. The development of gaseous and liquid hydrocarbons from polyethylene mix via hydropyropysis process is being studied. The hydropyrolytic process of converting LDPE, LLDPE, MDPE and HDPE type of polyethylene into useful gaseous and liquid hydrocarbon fuel provides a path to degrade waste polyethylene into small and very small chain products. The different fractionation of the extract obtained from the mother liquid was studied. The fraction ranging from temperature 150 °C to 400 °C has been studied to observe the diesel type fraction present in the liquid. The different blend study with the conventional diesel is being studied for property determination and end use. The chromatographic, distillate properties by simulated distillation analysis and mass spectra studies were carried out to understand the properties of the product. The blend study of the diesel type fuel from polyethylene with conventional diesel fuel provides a scope for its usefulness as feedstock in internal combustion engines and power generation.

Keywords: Polyethylene, Hydropyrolysis, Diesel, SIM-DIST, Mass spectroscopy, pour point.

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INTRODUCTION

Polyethylenes has become one of the major polymer material which world is using in each and every step of there lives. But the problem is to dispose these polyethylenes as waste after its use. The polyethylene does not degrade easily and hence become one of the major pollutants worldwide. About twenty percent of the total solid waste worldwide is polyethylenes. The conversion of the polyethylene into different hydrocarbon fuels either gaseous or liquid fuel by hydropyrolysis process is being studied from many years. The research is being carried out for different types of polyethylene such as LDPE (linear density polyethylene), **LLDPE** (linear low density polyethylene), MDPE (medium density polyethylene), and HDPE (high density polyethylene) by this process. The product obtained from this hydropyrolytic process were extracted and different components of these were separated. The different physical and chemical

properties of these products were studied and the different structural properties were observed.

Polyethylene made up of long chain carbon atoms and hydrogen atoms (-CH-)n along with some traces of oxygen, sulfur and nitrogen. These are generally chain linking in which large number of repeated units of monomers is bonded together.

$$nCH2 = CH2 \rightarrow [-CH2 - CH2 -] - n -$$
 (1)

Polyethylene, generally have very strong chemical bond that make it durable and resistant to natural activity. Polyethylene to hydrocarbon fuel is a de-polymerization process of converting waste polyethylene into useful hydrocarbon products. The process of converting the polyethylene into useful hydrocarbon product is followed by hydropyrolytic process [1]. The proportion of each fraction and there precise composition depends primarily on the nature of the plastic waste, but also on process condition.[2]

Molecular mass of polyethylene is high and it is possible that to improve the quality and performance and to reduce the cost, polyethylene may contain other substances. The polyethylenes contain some substances in the form of additives such as antioxidants, colorants, and other stabilizers. The oils derived from municipal solid polyethylene waste have high oxygen content due to the presence of cellulosic components in the waste. The oils derived from the hydropyrolysis of waste polyethylene materials tend to be chemically very complex due to the polymeric nature of the wastes and the range of potential primary and secondary reactions. Polyethylene waste hydropyrolytic oils contain hundreds of different chemical compounds, including organic acids, phenols, alcohols, aldehydes, ketones, The product oils derived furans. etc. hydropyrolysis of plastic waste produce end products which are based on the original polymer structure. Polymers of Polyethylene are much softened and can be extruded into required shapes.

The thermo chemical decomposition of organic material at elevated temperature in the absence of oxygen is studied. The thermal degradation of the polymers occurs by breaking C–C or C–H bonds. Degradation depends on the nature such as polar groups and structure, for example head-to-head or tail-to tail weak linkages, double bonds or branching points of the polymeric chain, on the ingredients in the polymer resin and on the type of the external stress. The different study of the diesel type product obtained from such process were visualized in this article.

METHODOLOGY

The polyethylene is hydropyrolysed to obtain gasious and mother liquid fractions. The mother liquid obtained from the process is finally fractionated in a column to obtain naphtha, middle distillate and heavy cut oil. The middle distillate generally contains the diesel type liquid which is extracted between temperature range 150 $^{\rm 0}{\rm C}$ to 400 $^{\rm 0}{\rm C}$.

Hydropyrolytic degradation of polyethylene is a complex series of chemical and thermal reactions to decompose or depolymerize organic material. Under oxygen free conditions with the presence of hydrogen and water medium. The liquid product is first extracted from the polyethylene waste. The polyethylene degradation mechanism is followed by random scission, end chain scission, de-propagation, intermolecular and intra-molecular hydrogen transfer β -scission, formation of branches and finally termination, either by bimolecular coupling or disproportion reaction.

The degradation process mainly follows the mechanism of chain scissions which is generally caused by increase in temperature and sheer stress in twin-skew extruders and hence decreases the molecular weight. Cross linking or chain branching have opposite effect to chain scission and are caused mainly by low sheer

stress and high residence time in a single skew extruders. A concurrence occurs between the molecular enlargement by chain branching and the decrease of molecular weight by chain scission. The thermal degradation of the polymers occurs by breaking C–C or C-H bonds. Degradation depends on the nature such as polar groups and structure, for example head to head or tail to tail weak linkages, double bonds or branching points of the polymeric chain, on the ingredients in the polymer resin and on the type of the external stress. In free radical mechanisms, the degradation of polymers generally involves the stages of initiation, propagation, chain transfer and termination. Initiation is the slowest reaction stage and generates macroradicals by chain scission or end chain breaking of the macromolecular backbone or by scission of the bonds with the pendant groups. The reactivity of the macroradicals, the R-CH2radical has a higher reactivity than the R2CH- and the mobility of the hydrogen atoms in the backbone, the tertiary hydrogen is more reactive than the secondary and primary one which strongly affects the mechanism of polymer degradation and decomposition [3].

This is also evident that the different component so produced may have undergone different reaction mechanism at different conditions.

During the thermal degradation of many polymers, other reactions may take place at the same time as the isomerization, cyclization, aromatization, recombination of species, etc [4].

A variety of products and applications can been observed by the thermal depolymerisation of polymeric materials through hydropyrolytic route.

Experimental

The fractional distillation of the recovered product from the depolymerization of the polyethylene mix is being carried out. The fraction ranging from 150 ⁰C to near about 400 ⁰C is being extracted from the mother liquid product. The product is then separated and blended with the conventional diesel fuel at different proportions range from 5%, 10%, 20%, 30%, 40%, 50%, 70%, 80%, and 90%. The blended product is then tested for different parameters observation. The temperature variation analysis through, Agilent, single column, split type with FID detector simulated distillation (SIM-DIST) and detailed hydrocarbon analyzer-front end (DHA-FE) instruments, were carried Simulated distillation (SIM-DIST) out. chromatographic technique which separates individual hydrocarbon components in the order of their boiling points. The separation is accomplished with a non-polar chromatography column using a gas chromatograph equipped with an oven and injector that can be temperature programmed. A flame ionization detector (FID) is used for detection and measurement of the hydrocarbon analytes. The result of SIM-DIST analysis provides a quantitative percent mass yield as a function of boiling point of the hydrocarbon components of the sample [5]. The detection of the temperature gradient with the ramp in temperature is observed. The fractional recovery of the product at different temperature ranges with respect to the mass % and volume% is observed. The change in different parameters such as cetane number, cetane index (calculated), flash point, kinematic viscosity, pour point, metal contents etc were observed. The different carbon number detection of the product on the basis of their boiling range is detected. The different blend study of the products with the conventional diesel fuel is described [6-8].

Hydropyrolytic degradation of polyethylene is a complex series of chemical and thermal reactions to decompose or depolymerize organic material. The complex nature of the diesel like fuel extracted from the polyethylene mix generally contains large amount of the paraffinic and olefinic components with some cyclic or naphthenic and aromatic components. With the increase in the paraffinic component and the isoparaffinic components in the diesel oil, the cetane number increases. But with the abundance of the linear fraction of the paraffinic groups the freezing point and the cloud point increases. With the increase in the

olefinic and the polyolefinic components the residual gum formation and the formation of unburned carbon residue increases. So in the hydropyrolysis process the hydrogen is added to reduce the formation of the olefinic radicals and large paraffinic components.

One of the most widely used characterization of the complex hydrocarbon mixture is the determination of the boiling point distribution. The different proportionate blend of the extracted diesel fuel with the conventional diesel fuel is analyzed to determine the characteristic property of the blended fuel for end use.

5% blend with diesel fuel

5% of diesel like component extracted from the polyethylene mix is mixed with the conventional diesel fuel and its properties were investigated.

The simulated distillation analysis of the blend is carried out and its boiling range at different temperature intervals is observed. The corresponding carbon number with respect to the boiling range though virtual is also identified for the component. The graph and the boiling range on the basis of volume and mass percentages are given below.

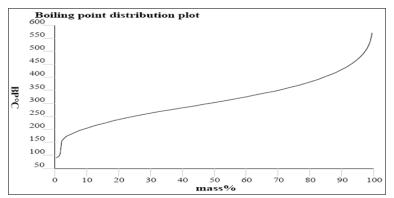


Fig-1: Boiling point distribution plot of 5% blend of diesel like component from polyethylene mix

The above plot describes the recovery or elution of the product with respect to the increase in temperature. The recovery at different intervals are observed on the basis of mass percentage.

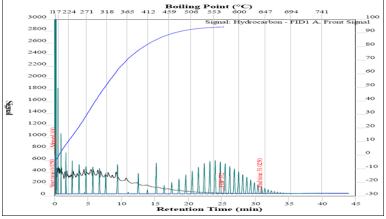


Fig-2: Boiling point distribution graph of the 5% volume blend of the diesel like component extracted from polyethylene mix on temperature versus retention time

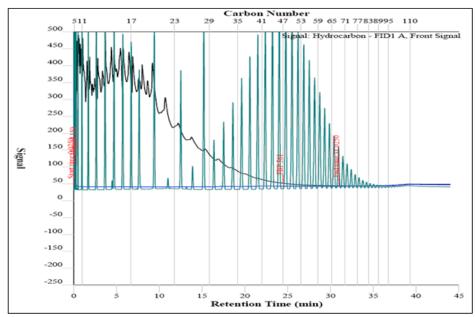


Fig-3: Carbon number distribution graph of the 5% blend of the diesel like component extracted from polyethylene mix

Table-1: Boiling point on the basis of volume percentage of the 5% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	153
5	206.8
10	223.2
15	234
20	245
25	254.4
30	2625
35	270
40	276
45	282
50	289.4
55	296
60	305.2
65	312
70	320.2
75	329
80	335.2
85	349.7
90	364.8
95	378.4
FBP	408.5

Some other characteristic property of the blended fuel were observed. The kinematic viscosity of the 5% blended fuel is found to be 2.515 cst and the pour point is observed to be 0.

10% blend with the diesel fuel

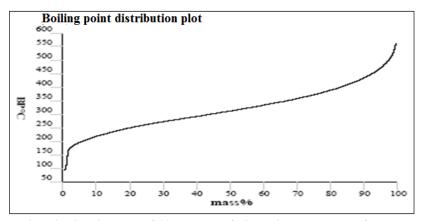


Fig-4: Boiling point distribution plot of 10% blend of diesel like component from polyethylene mix

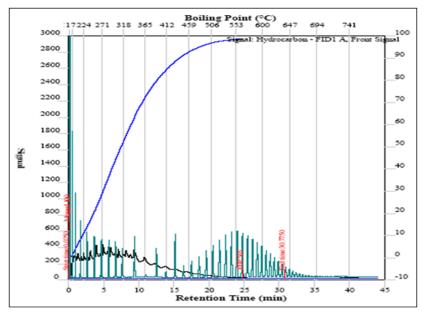


Fig-5: Boiling point distribution graph of the 10% volume blend of the diesel like component extracted from polyethylene mix on temperature versus retention time

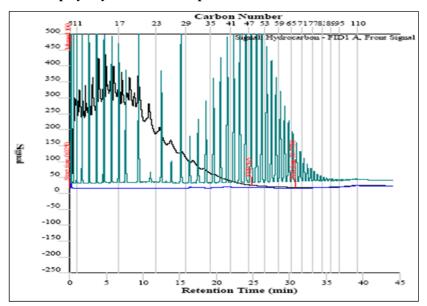


Fig-6: Carbon number distribution graph of the 10% blend of the diesel like component extracted from polyethylene mix

Table-2: Boiling point on the basis of volume percentage of the 10% blend of diesel like component with the conventional diesel fuel

COHVCI	itional diesel luci
Vol %	Boiling point (⁰ C)
IBP	153
5	207.4
10	225.2
15	234.5
20	247
25	255.4
30	2625
35	270
40	276.9
45	288
50	294.4
55	300.1
60	305.9
65	314
70	323.2
75	332
80	338.2
85	353.7
90	365.8
95	379.4
FBP	408.7

The kinematic viscosity of the 10% blended fuel is found to be 2.463 cst and the pour point is observed to be 0. **20%blend with the diesel fuel**

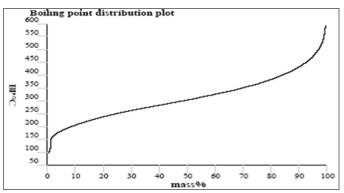


Fig-7: Boiling point distribution plot of 20% blend of diesel like component from polyethylene mix

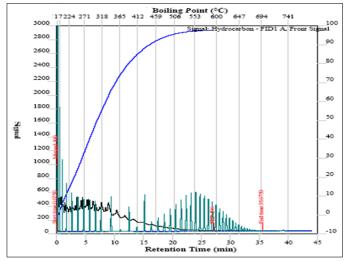


Fig-8: Boiling point distribution graph of the 20% volume blend of the diesel like component extracted from polyethylene mix on temperature versus retention time

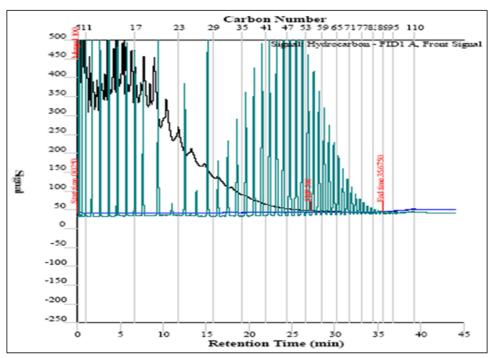


Fig-9: Carbon number distribution graph of the 20% blend of the diesel like component extracted from polyethylene mix

Table-3: Boiling point on the basis of volume percentage of the 20% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	153
5	206.4
10	224.2
15	235.5
20	246.2
25	253.4
30	2605
35	270.3
40	274.9
45	286.9
50	293.4
55	301.1
60	307.8
65	316
70	324.2
75	333.8
80	340.2
85	353
90	366.8
95	380.1
FBP	408.6

The kinematic viscosity of the 20% blended fuel is found to be 2.423cst and the pour point is observed to be +3.

30% blend with the diesel fuel

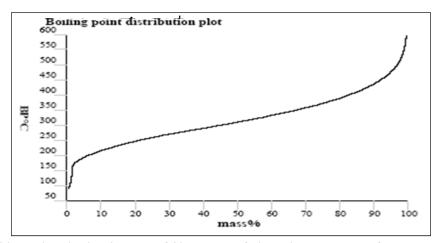


Fig-10: Boiling point distribution plot of 30% blend of diesel like component from polyethylene mix

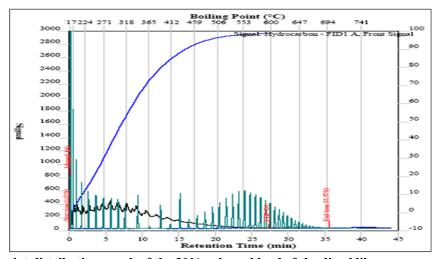


Fig-11: Boiling point distribution graph of the 30% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

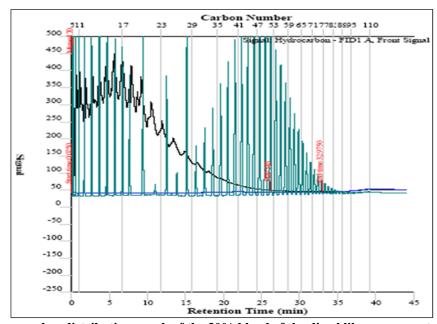


Fig-12: Carbon number distribution graph of the 30% blend of the diesel like component extracted from polyethylene mix

Table-4: Boiling point on the basis of volume percentage of the 30% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	152
5	201.4
10	224.2
15	233.5
20	242.4
25	251.7
30	2615
35	269.3
40	274.9
45	283.9
50	294.4
55	302.1
60	308.5
65	315.9
70	324.6
75	334
80	341.1
85	352
90	365.9
95	380.2
FBP	408.7

The kinematic viscosity of the 30% blended fuel is found to be 2.401 cst and the pour point is observed to be +3

40% blend with the diesel fuel

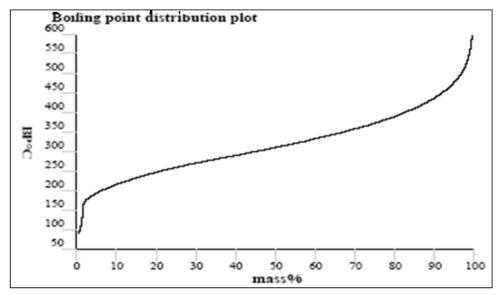


Fig-13: Boiling point distribution plot of 40% blend of diesel like component from polyethylene mix

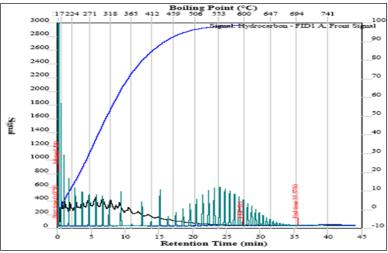


Fig-14: Boiling point distribution graph of the 40% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time.

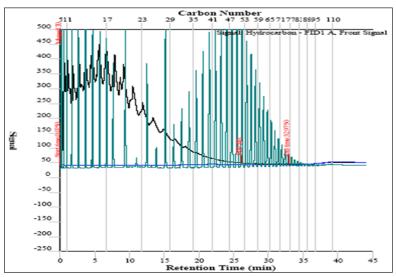


Fig-15: Carbon number distribution graph of the 40% blend of the diesel like component extracted from polyethylene mix

Table-5: Boiling point on the basis of volume percentage of the 40% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	142
5	190
10	203
15	219.3
20	232.2
25	250.1
30	261.2
35	271.2
40	277
45	284.7
50	296
55	303.5
60	310.1
65	318.2
70	327.2
75	336.1

80	343
85	353.1
90	368
95	379.6
FBP	409.1

The kinematic viscosity of the 40% blended fuel is found to be 2.391 cst and the pour point is observed to be +3

50% blend with the diesel fuel.

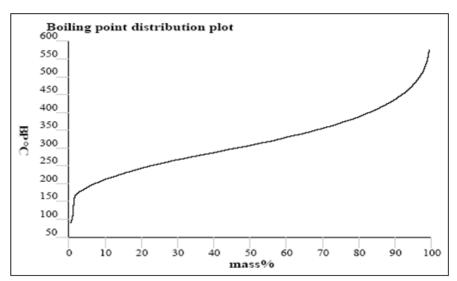


Fig-16: Boiling point distribution plot of 50% blend of diesel like component from polyethylene mix

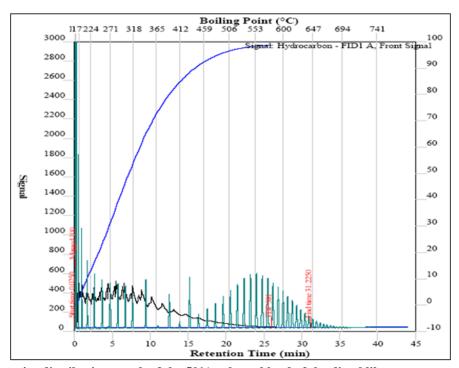


Fig-17: Boiling point distribution graph of the 50% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

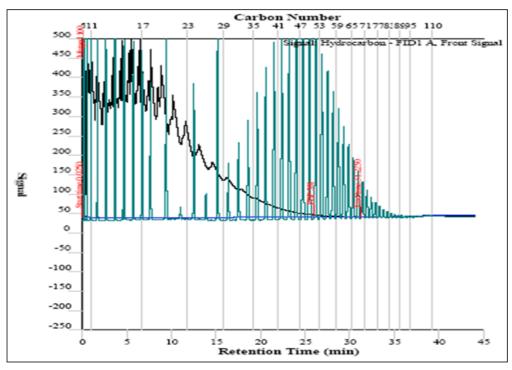


Fig-18: Carbon number distribution graph of the 50% blend of the diesel like component extracted from polyethylene mix

Table-6: Boiling point on the basis of volume percentage of the 50% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	141.3
5	189.3
10	203
15	221.1
20	234.2
25	252.1
30	261.1
35	273.2
40	279
45	285.7
50	298
55	306.5
60	313.1
65	321.2
70	329.2
75	339.1
80	348
85	358.1
90	372
95	382.6
FBP	409.1

The kinematic viscosity of the 5% blended fuel is found to be 2.356cst and the pour point is observed to be +3.

60% blend with the diesel fuel.

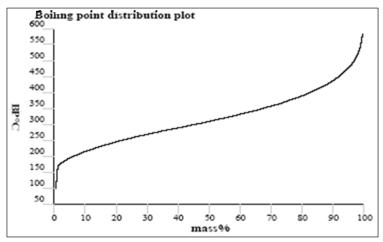


Fig-19: Boiling point distribution plot of 60% blend of diesel like component from polyethylene mix

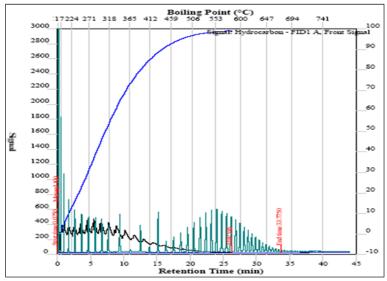


Fig-20: Boiling point distribution graph of the 60% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

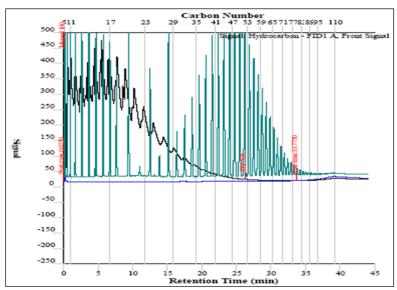


Fig-21: Carbon number distribution graph of the 60% blend of the diesel like component extracted from polyethylene mix

Table-7: Boiling point on the basis of volume percentage of the 60% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	141.6
5	168.3
10	179
15	195.1
20	208.9
25	219.1
30	229.3
35	238.2
40	248.3
45	258.7
50	269
55	276.5
60	389.2
65	300.2
70	316.2
75	338.1
80	349.2
85	359.1
90	369.9
95	381.6
FBP	410.4

The kinematic viscosity of the 60% blended fuel is found to be 2.312 cst and the pour point is observed to be +3

70% blend with the diesel fuel.

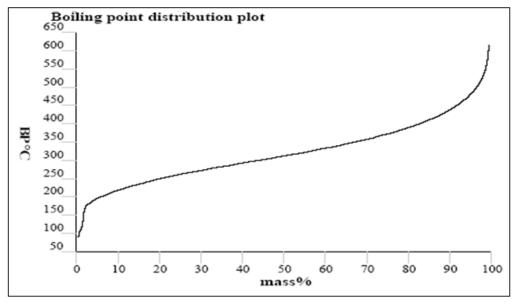


Fig-22: Boiling point distribution plot of 70% blend of diesel like component from polyethylene mix

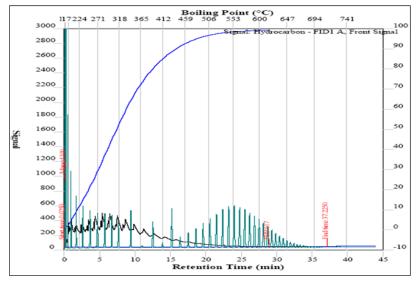


Fig-23: Boiling point distribution graph of the 70% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

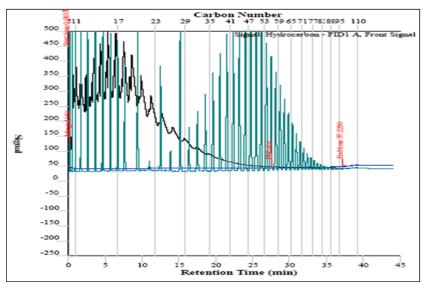


Fig-24: Carbon number distribution graph of the 70% blend of the diesel like component extracted from polyethylene mix

Table-8: Boiling point on the basis of volume percentage of the 70% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	141.4
5	167.3
10	178.5
15	194.5
20	207.8
25	218.1
30	226.4
35	239.2
40	247.3
45	256.8
50	268.5
55	277.4
60	387.2
65	301.3

70	315.3
75	337.4
80	348.4
85	358.9
90	367.8
95	380.4
FBP	410.7

The kinematic viscosity of the 70% blended fuel is found to be 2.215cst and the pour point is observed to be +3.

80% blend with the diesel fuel.

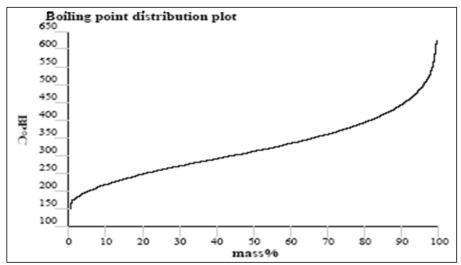


Fig-25: Boiling point distribution plot of 80% blend of diesel like component from polyethylene mix

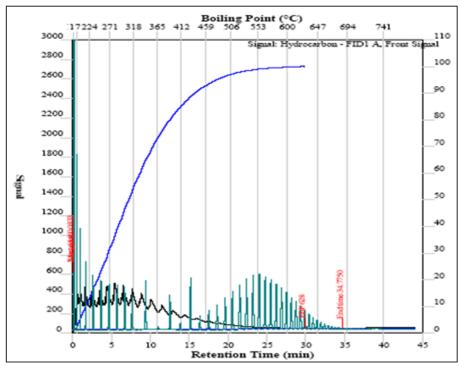


Fig-26: Boiling point distribution graph of the 80% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

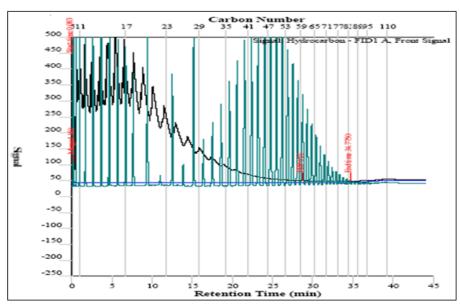


Fig-27: Carbon number distribution graph of the 80% blend of the diesel like component extracted from polyethylene mix

Table-9: Boiling point on the basis of volume percentage of the 80% blend of diesel like component with the conventional diesel fuel

Vol %	Boiling point (⁰ C)
IBP	142.1
5	160.3
10	167.5
15	182.5
20	191.8
25	199.1
30	205.4
35	216.2
40	228.4
45	239.8
50	251.2
55	261.2
60	372.2
65	385.2
70	309.3
75	324.4
80	338.9
85	356.9
90	367.8
95	373.4
FBP	410.3

The kinematic viscosity of the 80% blended fuel is found to be 2.202cst and the pour point is observed to be +3

90%blend with the diesel fuel.

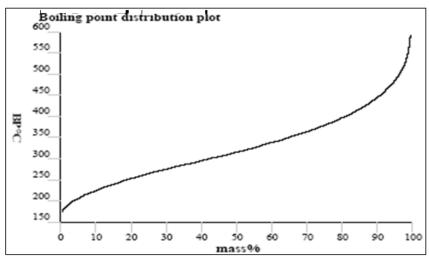


Fig-28: Boiling point distribution plot of 90% blend of diesel like component from polyethylene mix

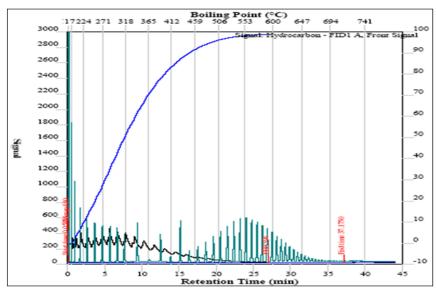


Fig-29: Boiling point distribution graph of the 90% volume blend of the diesel like component extracted from polyethylene mix by temperature versus retention time

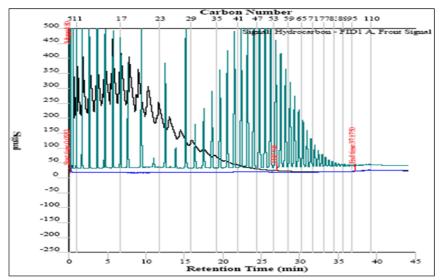


Fig-30: Carbon number distribution graph of the 90% blend of the diesel like component extracted from polyethylene mix

Table-10: Boiling point on the basis of volume percentage of the 90% blend of diesel like component with the conventional diesel fuel

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Vol %	Boiling point (⁰ C)
IBP	140.9
5	156.6
10	161.3
15	176.5
20	184.9
25	195.2
30	203.4
35	214.5
40	227.4
45	239.8
50	256.2
55	266.2
60	375.2
65	387.2
70	309.3
75	328.4
80	336.9
85	345.9
90	354.8
95	367.6
FBP	410.3

The kinematic viscosity of the 90% blended fuel is found to be 2.168 cst and the pour point is observed to be +3.

The above blend study of the diesel like component with the conventional diesel fuel provides information about the characteristics property of the fuel to be used as diesel fuel in the automobiles and diesel engines. Distillation curves give the amount of a fuel sample that is evaporated at atmospheric pressure at each temperature range, when the temperature is increased gradually. Distillation characteristics illustrate how fuel is evaporated when it is sprayed into the combustion chamber of a diesel engine. Some fuel fractions which boil at low temperatures are needed for engine start up, while fractions boiling at too high temperatures may not combust completely forming engine deposits [9].

The experimental study described above provides the evidence of the different diesel fraction recovered by the blend of the conventional diesel fuel with the extracted diesel fuel from polyethylene mix.

Cetane analysis of blended diesel fuel

The cetane number is a measure of how readily the fuel starts to burn (auto ignites) under diesel engine chamber. A fuel with a high cetane number starts to burn shortly after it is injected into the cylinder, therefore it has a short ignition delay period. Conversely, a fuel with a low cetane number resists auto ignition and has a longer ignition delay period. In a diesel engine, fuel ignites spontaneously, shortly after injection begins. During this delay, the fuel vaporizes and mixes with the air in the combustion chamber.

Combustion causes a rapid heat release and a rapid rise of combustion chamber pressure. The rapid rise in pressure is responsible for the knock that is very audible in some diesel engines. By increasing the cetane number of the fuel, the knock intensity is decreased by the shortened ignition delay. Fuels with high cetane numbers ignite before most of the fuel is injected into the combustion chamber. The rates of heat release and pressure rise are then controlled primarily by the rate of injection and fuel air mixture and smoother engine operation results [10]. Cetane analysis of the blended diesel fuel is carried out to understand the combustion characteristic and the knocking property of the fuels. The different blended products are analyzed using cooperative fuel research (CFR) engine. The CFR engine is based on a single cylinder compression ignition engine which comprises of the wheel having 900 rotations per minute (RPM) [11].

The fuel is injected in the engine and on the basis of the ignition advance and ignition delay with the use of the drum adjustment the cetane number is calculated. The cetane number is the knocking characteristic of the fuel which varies with the different hydrocarbon structures. Normal paraffin has high cetane numbers which increases with molecular weight. Iso-paraffins have wide range of cetane numbers, from about 10-90. Molecules with many short side chains have low cetane numbers, whereas those with one side chain of four or more carbon have high cetane numbers. Higher molecular weight molecules with one long side

chain have high cetane numbers. Lower molecular weight molecules with short side chain have low cetane numbers.

The aromatic component has low cetane number. The blended products were analyzed and the different cetane numbers observed are described below.

Table-11: Cetane number of different blended products of diesel like component with the conventional diesel fuel

Blend percentage with conventional diesel fuel (in	Cetane number
%)	
5	52.1
10	53.2
20	54.3
30	55.8
40	56.6
50	57.7
60	60.1
70	64.7
80	70.4
90	76.9

The cetane number of the conventional diesel fuel is observed to be 51 cetane. The diesel with cetane number 51 is blended with the extracted diesel like component extracted from the mother liquid by hydropyrolytic depolymerization of polyethylene mix in different proportions. The different cetane values were observed. The cetane number of the blended products

from 5% to 90 % is found to be in the increasing order. The graph for the cetane number at different intervals is described below. This reveals that with the increase in the blended products the increasing paraffinic component present in the diesel like component increases and hence the cetane number of the blended products also increases gradually.

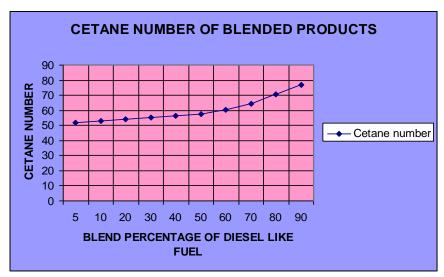


Fig-31: Graph showing the blended diesel like product with the conventional diesel fuel at different proportions

The above graph shows the increase in the cetane number value with the increase in the blending proportions of the fuel. The cetane number value of the above products is carried out using the bracketing methods. Two known valued cetane materials were used to bracket the desired fuel for determination of the cetane number. These known valued materials are generally represented as high reference fuel (HRF) and low reference fuel (LRF). These reference fuels are bracketed with the desired fuel (the different blend proportions of the fuel) for the determination of the cetane value. The low reference fuel, high reference fuel and the blended samples were injected in the

combustion chamber one by one and the ignition delay and the ignition advance criteria were adjusted accordingly.

On the basis of the combustion characteristics the desired cetane value for the different blended products were observed and estimated. The cetane number and the different properties of the blended fuels at different proportions reveal that the fuel can be used as a conventional fuel with proper blending.

CONCLUSION

The article provides information about the different blending properties of the extracted diesel from polyethylene mix with component conventional diesel component. The blended components have different distinct properties which makes the product to be used for end user. The high cetane value of the component without or very less change in the pour point, flash point and the viscosity property of the blended product makes it more user and engine friendly. The simulated distillation analysis of the blended products provides information about the different boiling point distribution of the products. It was observed that as much as 30 percent of the extracted diesel cut component from the polyethylene mix can be blended to the conventional diesel for use as commercial diesel fuel. This will save about 30 percent of the total diesel production cost by maintaining sustainable and greener environment from hazardous polyethylene waste.

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