Production of biodiesel from groundnut (*Arachis hypogea,* L.) oil

O.O. Oniya[1*], A.I. Bamgboye[2]

(1. *Department of Agricultural Engineering, Ladoke Akintola University of Technology, P.M.B. 4000, Ogbomoso, Oyo State, Nigeria;*
2. *Department of Agricultural Engineering, University of Ibadan, Ibadan, Nigeria*)

Abstract: Biodiesel is an environmental-friendly substitute for fossil fuel. Research on the production of biodiesel from vegetable oils has concentrated on Jatropha, soybean, palm kernel, sunflower and rapeseed oils with scarce information on groundnut oils. This study focussed on the production of biodiesel from groundnut oil. Oil was mechanically extracted from groundnut seeds that were bought from *Sabo* market in Ogbomoso, Nigeria. The extracted oil was trans-esterized with ethanol using potassium hydroxide as catalyst in a two-step trans-esterification process to yield ethyl esters and glycerol. The fuel properties of groundnut oil and its ethyl ester were determined according to American Society for Testing Materials (ASTM) standard methods and compared with that of Automotive Gas Oil (AGO). The results showed that groundnut seeds gave oil yield of 45.3%, while trans-esterification yielded 86.8% groundnut oil ethyl ester on a volume basis. At 40ºC, higher viscosity of 39.2 mm^2/s was obtained for raw groundnut oil than 7.60 mm^2/s obtained for groundnut oil ethyl ester. At 15ºC, specific gravity of raw groundnut oil and its ethyl ester were 0.9 (1.047 times that of AGO) and 0.85 (1.012 times that of AGO) respectively and are within limit specified by international standards. The biofuels contained lower amounts of sulphur (9.73% for groundnut oil ethyl ester and 12.8% for raw groundnut oil) than the reference AGO which was 61.8%. Higher pour (4ºC and 3ºC) points, cloud (7ºC and 8ºC) points and flash (200ºC and >280ºC) points were obtained for groundnut oil ethyl ester and the raw groundnut oil respectively compared to -16ºC, -12ºC and 74ºC respectively obtained for AGO. The fatty acid profile of the groundnut oil reveals 75.03% unsaturated fatty acids in the oil composition. Groundnut oil ethyl ester was found to have better fuel quality than raw groundnut oil and it has potentials to fuel a diesel engine.

Keywords: groundnut oil, automotive gas oil, transesterification, ethyl esters, biodiesel, fuel, diesel engine, Nigeria

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1 Introduction

Biofuels have become more attractive recently as an alternative fuel because of its renewability and positive environmental benefits. It burns up to 75% cleaner than fossil fuels. It substantially reduces unburned hydrocarbons, carbon monoxide and particulate matter in exhaust fumes which cause cancer. Also, it is a much better lubricant than fossil fuels and extends engine life (Hobbs, 2003; Faupel and Kurki, 2002; Elsbett and Bialkowsky, 2003). Biofuels increase safety in storage and transport because they are non-toxic and biodegradable (Hobbs, 2003; Faupel and Kurki, 2002; Elsbett and Bialkowsky, 2003). Van Gerpen et al. (1996) had found that the cetane numbers of biodiesel depend on the fatty acids profile of the original oil or fat in which it is produced. He found that the longer the fatty acid carbon chains and the higher the unsaturation, the higher the cetane number. Biofuels have been described as safe fuels, because the flash point is more than 100°F higher than that of diesel and was given as 160°C which showed that it can be classified as a non-flammable liquid; the toxicity is at least 15 times less than diesel (Peterson et al., 1994). The flash point of petroleum diesel was given as 64°C and that of gasoline as 45°C (Peterson et al., 1994).

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* **Corresponding author: O. O. ONIYA**, Department of Agricultural Engineering, Ladoke Akintola University of Technology, P.M.B. 4000,Ogbomoso, Oyo State, Nigeria. Email: toyinprayer@yahoo.com.

Rakopoulos et al. (2006); Faupel and Kurki (2002); Mohammed et al. (2002) gave a number of disadvantages encountered when using biodiesel in compression – ignition engine. They observed that biodiesel fuels have higher viscosity, higher pour point, lower heating value and lower volatility than petroleum diesel fuel. Also, the oxidation stability of biodiesel fuels was found to be lower and hygroscopic. As solvents they cause corrosion of component, attacking some plastic materials used for seals, hoses, paints and coatings. They showed increased dilution and polymerization of engine sump oil, thus requiring more frequent oil changes. Furthermore, the use of vegetable oil for fuel causes competition with the use of vegetable oil for food and it has higher per gallon cost than petroleum diesel in the current market. Ahouissoussi (1995) has noted that although biodiesel have higher total costs than diesel fuel, they have the potential to compete with compressed natural gas and methanol as fuels for urban transits buses. Also, it is noteworthy that Krawczyk (1996) identified biodiesel as a possible replacement to fossil's fuels as the world's primary energy source. Biodiesel could be obtained from an array of agricultural produce such as soybean, groundnut etc.

Groundnut (*Arachis hypogea, L.*) belongs to the Leguminosae family and it propagates itself by self – pollination. Nigeria is the third largest producer of groundnut after China and India (SATRENDS, 2001). In 1994, 1,453,000 tons of groundnut were produced in the country. While in 1998, 2,227,000 tons of the crops were produced in the country (CBN, 1998).

The Nigerian oil mill company in Kano, Kano State, Nigeria crushes between 100–120 tons of groundnut seeds a day during their oil production season (SATRENDS, 2001). In addition, there are numerous oil mills (both large-scale and low-scale industries) which ensure the production of groundnut oil throughout the year. Therefore, the availability and abundance of the oil makes it a candidate for being used for the production of biodiesel.

The fuel quality and performance of biodiesel are affected by the raw materials used, the method of production, amount of biofuel blended with petroleum diesel, type of esterification process, etc. (Moreno et al. 1999). Among the crops grown in the tropics, considerable work has been done using vegetable oils such as soybean oil, palm oil, palm kernel oil, sunflower oil, rape seed oil, *Jatropha* oil, etc. (Kinast, 2003; Darnoko and Cheryan, 2000; Alamu et al. 2007; Best, 2006). No work has been reported on the production of groundnut oil ethyl ester as a fuel for diesel engine. In this work, Nigerian groundnut oil was transesterified. The raw oil and its ethyl ester were evaluated as fuel for diesel engines.

2 Materials and methods2.1 Sample preparation and experimental procedures

Groundnut seeds bought from a market in Ogbomoso, Oyo State, Nigeria were used for this work. The seeds were processed using mechanical extraction method. The extracted oil was transesterified to produce alcohol esters using a closed reactor in a two-step transeterification process as developed by Peterson et al. (1996) and Saifuddin and Chua (2004). The fuel properties of raw groundnut oil and its alcohol esters were determined according to the method elaborated in American Society for Testing Materials (ASTM D40, D93, D97, D445, D1298, D2500, A6584) standards. Also, petroleum diesel fuel used as reference was bought from a local filling station in Ogbomoso and the fuel properties were also determined using the method outlined in ASTM D40, D93, D97, D445, D1298, D2500, A6584 standards.

Ethanol was chosen for this work as the alcohol to trans-esterify groundnut oil because it is renewable and derived from locally available agricultural products. Also, it is biologically less objectionable in the environment. Potassium hydroxide was used as the catalyst in the reaction because it was cheaper and the performance was better than other catalysts available.

Three operations are performed in the production of the groundnut oil ethyl ester as shown in Figure 1. These were transesterification, phase separation and washing.

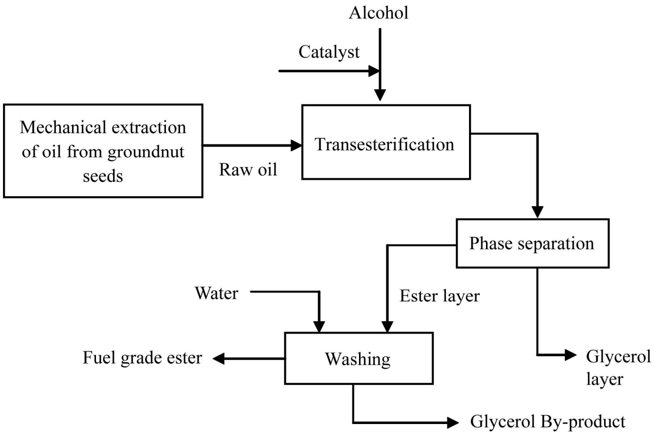

Figure 1 Detailed process flowsheet for the preparation of biodiesel fuel

2.1 The mechanism of the transesterification process

The reaction temperature of 60ºC was selected as suggested by Alamu et al. (2007). Potassium hydroxide was added to ethanol to form potassium ethoxide. The ethanol-KOH mixture (potassium ethoxide) was poured into the groundnut oil in a transesterification reactor, and the following transesterification reaction occured (a hypothetical formula for plant oil suggested by Peterson et al. (1996) and presented in Equation (1). The mixture of potassium hydroxide and ethanol in the flask was stirred vigorously. The ethanol catalyst mixture was poured into the oil in the main reactor and stirred rapidly.

O
H(2)C-O-C-C(19)H(35) H(2)C-OH
O O
H-C-O-C-C(19)H(35) + 3C(2)H(5)OH......3C(19)H(35)COC(2)H(5) + H-C-OH (1)
O
H(2)C-O-C-C(19)H(35) H(2)C-OH
(Hypothetical Formula) (Hypothetical Formula)

1 mol of triglyceride (plant oil) + 3 moles of ethanol 3 moles of ethyl ester + 1 mol of glycerol.

It was deduced from Equation (1) that the transesterification reaction is reversible and thus an excess alcohol is usually used to force the equilibrium to the product side. Therefore, ethanol was added at 65% stoichometric excess, or a molar ratio of 5.0:1 (ethanol to oil).

The weight of potassium hydroxide used as catalyst during transesterification was calculated from the following formula as recommended by Peterson et al. (1996).

$$KOH = 0.013 \times GO \quad (2)$$

where, GO = the desired volume of groundnut oil processed in litres; KOH = Weight of potassium hydroxide required in kilogram.

Therefore, potassium hydroxide was added at 1.5% of the weight of oil.

Phase separation of the products of transesterification was achieved following the procedure developed by Peterson et al., (1996). After initial stirring for one hour, water was added at fifteen percent of the initial volume of oil used in the reaction and the entire content was re-stirred for ten minutes. The mixture was allowed to settle for 48 h. After settling and completion of separation, glycerol which is the heavier liquid collected at the bottom, while the ester product was at the top. The glycerol layer was drained off and the ester layer remained as shown in Figure 2.

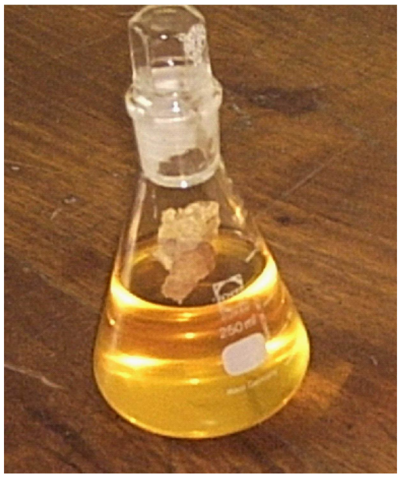

Figure 2 Pure biodiesel from groundnut oil

2.2 The washing process

The method used for washing consists of two steps developed by Peterson et al. (1996), is described as follows:

a) The glycerol layer was re-mixed with the ester layer after initial settling has occurred, then 15% water was added and the entire mixtures were restirred for 10 min and allowed to settle for 48 h.

b) The ester product was washed after draining off the glycerol layer. The ester was washed with water at about 30% of the ester volume. The water was stirred into the ester with mechanical stirring using a blender. After 10 min, the stirring was stopped, and the water was allowed to settle out for two days. At this point, the process was complete and the crystal clear product was the groundnut oil ethyl ester.

2.3 Determination of the yield of esters

The yield of the ethyl esters, Y produced was calculated thus:

$$Y = \frac{V_e}{V_r} \times 100\% \tag{3}$$

where, Y = Yield of the ethyl esters, %; V_e = Volume ethyl esters produced in litres; V_r = Volume raw oil used in litres.

2.4 Determination of the fuel properties of groundnut oil and its ethyl esters

The kinematic viscosity of the fuel samples was determined by the method in ASTM D445 using Cannon-Fenske Capillary Viscometer tube. The specific gravity of the fuel samples was determined by means of pyconometer according to the method described in ASTM D1298. The flash point of the fuel samples was determined by heating a sample of the fuel in a stirred container and passing a flame over the surface of the liquid according to the method described in ASTM D93 by using the Pensky Martens flash point closed apparatus.

As described in ASTM D97, the pour point of the fuel samples was determined by cooling the sample at a specified rate and the sample was examined at 3ºC intervals for flow. The lowest temperature at which sample movement was observed was noted as the pour point. Also, as described in ASTM D2500, the cloud point of the fuel sample was determined by visually inspecting for a haze in the normally clear fuel, while the fuel was cooled under carefully controlled conditions. The percentage free fatty acid composition of groundnut oil was determined using Gas Chromatography (GC) according to method described in ASTM A6584. The heating value of fuel samples was determined by the method in ASTM D40. A Gallenkemp ballistic bomb calorimeter was used to determine the heating value. A regression equation developed by Bamgboye and Hansen (2008) was used to determine the cetane number of groundnut biodiesel. The saponification value, iodine value, pH value, ash content, sulphur content and carbon content of the oil samples were determined according to the method described in the American Oil Chemists' Society (AOCS) official and tentative methods. The detail procedures of all these analytical tests were outlined in Oniya (2010).

3 Results and discussion

The results obtained in this work include the yield obtained for the extraction of groundnut oil; yield of groundnut ethyl esters after transesterification and fuel properties of groundnut oil and its ethyl esters.

3.1 Groundnut oil yield

The results of extraction of oil from groundnut seeds by mechanical method are presented in Table 1. Oil yield of 45.3% was obtained by mechanical extraction.

Table 1 Groundnut oil extraction

Experiment No	Mass of milled groundnut sample, M_1 /g	Mass of groundnut oil, M_{lo} /g	Percentage oil yield, $\frac{M_{lo}}{M_1} \times 100\%$/%
1	7900.0	3580.0	45.3
2	8851.7	4027.5	45.5
3	2106.4	950.0	45.1
		Average	45.3

3.2 Transesterification result

The trans-esterification process yielded 86.8% groundnut oil ethyl ester on a volume basis (Table 2). The yield of ethyl ester was quite high and similar to the value obtained by Alamu et al. (2007) who worked on alkali-catalysed trans-esterification of palm kernel oil, and the process yielded 95.80% palm kernel oil ethyl ester on weight basis.

Table 2 The results of transesterification experiment

Experiment No	Volume of groundnut oil/mL	Volume of groundnut oil ethyl ester/mL	Yield of esters/%
1	500	410	82.0
2	1000	910	91.0
3	2000	1750	87.5
Average			86.8

3.3 The fatty acid profile of groundnut oil

The fatty acid profile of groundnut oil is presented in Table 3. It was observed that groundnut oil contains 11.69% palmitic acid, 41.64% oleic acid and 32.04% linoleic acid, while the remaining acids are in negligible percentages. The result showed that groundnut oil comprised of 75.03% unsaturated fatty acid with mainly 41.64% oleic acid and 32.04% linoleic acid.

Table 3 The fatty acid profile of groundnut oil

Component	Formula	Percentage composition	Name	Type
C 16:0	$C_{16}H_{32}O_2$	11.69	Palmitic acid	Saturated.
C 18:0	$C_{18}H_{36}O_2$	3.84	Stearic acid	Saturated.
C 18:1	$C_{18}H_{34}O_2$	41.64	Oleic acid	Unsaturated.
C 18:1	$C_{18}H_{34}O_2$	0.46	Oleic acid	Unsaturated.
C 18:2	$C_{18}H_{32}O_2$	32.04	Linoleic acid	Unsaturated.
C 20:0	$C_{20}H_{40}O_2$	1.84	Arachidic acid	Saturated.
C 20:1	$C_{20}H_{38}O_2$	0.89	Arachidoleic acid	Unsaturated.
C 22:0	$C_{22}H_{44}O_2$	4.05	Behenic fatty acid	Saturated.
C 24:0	$C_{24}H_{48}O_2$	1.53	Saturated fatty acid	Saturated.

The composition of groundnut oil was similar to rapeseed oil (64.1% oleic acid, 22.5% linoleic acid and others), *Jatropha* oil (42.1% oleic acid, 30.9% linoleic acid and others), sunflower oil (44.0% oleic acid, 10.7% linoleic acid and others), palm oil (43.1% oleic acid, 10.4% linoleic acid and others), neem oil (41.3% oleic acid, 16.4% linoleic acid and others) and karanja oil (53.2% oleic acid, 19.1% linoleic acid and others) which had more oleic acid than linoleic acid and were found suitable as sources of biodiesel fuels. Based on the fatty acid profile, groundnut oil is expected to provide a suitable source of biodiesel fuels (Van Gerpen, 1996; Rakopoulos et al. 2006; Kinast, 2003).

3.4 Fuel properties

The results of the fuel properties of raw groundnut oil, its ethyl esters and AGO used are presented in Table 4.

A viscosity of 39.2 mm^2/s and 7.60 mm^2/s were obtained for raw groundnut oil and the groundnut oil ethyl ester, respectively. Groundnut oil ethyl ester showed 80.61% reduction in viscosity thus enhancing its fluidity in diesel engine. The viscosity of groundnut oil ethyl ester was close to the reference AGO and fell within the 4-D grade diesel fuel. This showed that the groundnut oil ethyl ester was sufficiently viscous as fuel for diesel engines and that real spray would generate across the combustion chamber and this would be properly mixed with air. Kinematic viscosities of groundnut oil ethyl ester with AGO at various temperatures are as shown in Table 5. The viscosity of the biodiesel fuel decreased with increase in temperature as recorded for reference AGO, which was similar to the results obtained by Alamu et al. (2007) for biodiesel produced from palm kernel oil and the result obtained by Ajav and Akingbehin (2002) for biodiesel produced from ethanol.

Table 4 Fuel properties of pure diesel, groundnut oil ethyl ester, raw groundnut oil

Fuel properties	Ago	Groundnut oil ethyl ester	Raw groundnut oil
Viscosity at 40℃ (Cst)	2.95	7.60	39.20
Heating Value (MJ/L)	44.68	30.07	37.40
pH	2.80	2.90	3.40
Specific Gravity at 15℃	0.86	0.85	0.90
Cloud Point (℃)	-12	7	8
Pour Point (℃)	-16	4	3
Ash Content (%)	0.12	0.01	0.01
Flash Point (℃)	74	200	>280
Sulphur Content (%)	61.80	9.73	12.80
Carbon Content (%)	13.40	10.40	24.80
Iodine Value (wijis)	0.21	0.34	0.41
Peroxide value (meq/KOH)	0.14	0.13	0.13
SaponificationValue (MgKOH/g)	0.17	0.17	0.17
Free Fatty Acid (g/100 g)	8	7	11

Table 5 Kinematic viscosity of groundnut oil ethyl ester and AGO at various temperatures in mm^2/s

Samples	27ºC	40ºC	60ºC	100ºC
AGO	5.7	5.55	3.2	2.1
Groundnut oil ethyl ester	23.76	16.8	10.8	4.9

At 15ºC, specific gravity of raw groundnut oil and groundnut oil ethyl ester were 0.9 (1.047 times that of AGO) and 0.85 (1.012 times that of AGO) respectively. The low value of the specific gravity of the groundnut oil and its ethyl ester indicated good ignition property (Clark, 1998). This implies that groundnut oil and its ethyl ester have good combustion characteristics. The specific gravity values obtained for both groundnut oil and its ethyl ester fell within the limit specified by various international standards EN14214 (Europe), ONC1191 (Austria), CSN656507 (Czech Rebublic), Journal Officiel (France), DINV51606 (Germany), UN110635 (Italy) and

SS155436 (Sweden) standards which range from 0.86-0.9, 0.85-0.89, 0.87-0.89, 0.87-0.9, 0.875-0.9, 0.86-0.9 and 0.87-0.9 respectively for biodiesel fuels.

The heating value of groundnut oil ethyl ester was 30.07 MJ/L, which was lower than 37.40 MJ/L obtained for raw groundnut oil. This showed that the trans-esterification reduces the heating value and therefore the energy content of the vegetable oil. The reduction in the energy content was due to the fact that the carbon content of groundnut oil reduced considerably after transesterification from 24.8% to 10.4%. However, the heating values of groundnut oil and its ethyl ester were lower than that of AGO obtained as 44.68 MJ/L. Also, the heating values of the samples were quite high and close to the values obtained for soybean oil (34.7 MJ/L), *Jatropha* oil (34.7 MJ/L) and tigernut oil (34.6 MJ/L) which have been found as useful biofuels for diesel engines (Barminas et al. 2001). Therefore, groundnut oil and its ethyl ester have potentials to power a diesel engine.

The biofuels contained lower amounts of sulphur (9.73% for groundnut oil ethyl ester and 12.8% for raw groundnut oil) than the reference AGO which was 61.8%. Therefore, SO_x emissions are expected to be considerably reduced in diesel engines using the biofuels.

Higher pour (4ºC and 3ºC), cloud (7ºC and 8ºC) and flash (200ºC and >280ºC) were obtained for groundnut oil ethyl ester and the raw groundnut oil respectively compared to -16ºC, -12ºC and 74ºC respectively obtained for AGO. Also, the high flash point (more than 100ºC) of the biofuel ensures safe storage and safe transportation free from fire hazards. The higher cloud and pour points of the biofuel than the reference diesel fuel may involve some complications for their use in diesel engine during cold weather.

The same value (0.01) was obtained for the ash content of groundnut oil and its ethyl esters. The ash contents of groundnut oil and its ethyl esters were lower compared to AGO obtained as 0.12. Since the ash content is a measure of the amount of metal contained in the fuel, therefore this result indicated that the use of groundnut biofuels would reduce injector tip plugging, combustion deposits and injector system wear compared to AGO which had higher ash content. The use of the biodiesel fuels would not constitute a corrosion problem in the injection system and pressure chamber of a diesel engine. The results were consistent with the values of ash contents obtained for *Jatropha* oil, rapeseed methyl ester, sunflower methyl ester and *Jatropha* methyl ester obtained as 0.03, 0.007, 0.004 and 0.013 respectively (Moreno et al. 1999; Rahman et al. 2010; Labeckas et al. 2006).

The same saponification value (0.17 mgKOH/g) was obtained for groundnut oil, its ethyl ester and AGO. These results showed that groundnut biodiesel and AGO would behave similarly in the same chemical reaction.

The pH value of raw groundnut oil and its ethyl ester were found to be 3.4 and 2.9 respectively, which implied that that raw groundnut oil was not as acidic compared to its ethyl ester. Also, the pH of all the biofuels were higher but are close to that of AGO obtained as 2.8. The low pH value was due to the sulphur content.

The iodine value of groundnut oil obtained as 0.41 wijis was higher compared to its ethyl ester which was obtained as 0.34 wijis. This result indicated that the process of transesterification reduced the iodine value which is a measure of the stability of the biofuels during storage. Therefore, the ethyl ester was more stable than the raw oil. This was because the lower the iodine value, the more stable the fuel.

The cetane number of groundnut oil ester was determined as 53.8. The cetane number of pure linoleic acid was reported as 36.8, while that of oleic acid was reported as 57.2 (Bamgboye and Hansen, 2008). Therefore, the cetane number of groundnut biodiesel was within the range of the cetane number of dominating fatty acid constituents. This agreed with the findings of Bamgboye and Hansen (2008) who reported that the cetane numbers of esters of soybean, rapeseed, sunflower, cottonseed, peanut, palm oil, lard, tallow and canola oils were within the range of the cetane number of the dominating fatty acid constituents. The cetane number of groundnut biodiesel was also close to the values reported by Bamgboye and Hansen (2008) and Moreno et al., (1999) for esters of soybean oil (45 - 60), rapeseed oil (44 – 59), sunflower oil (50 – 61.2), cottonseed oil (45 –

55), peanut oil (54), palm oil (58 – 70), lard (63.6), tallow (58 – 64.8) and canola oil (53.9 – 55). The cetane number of groundnut biodiesel obtained as 53.8 agreed with the biodiesel standard of 49 minimum specified by the Technical Standard of the European Union.

4 Conclusions

From the results of transesterification of groundnut oil, biofuel characterisation of raw groundnut oil and groundnut oil ethyl ester carried out, the following conclusions were drawn;

- Transesterification will enhance fluidity of groundnut oil ethyl ester in diesel engine and reduce the heating value and therefore the energy content of the vegetable oil. Also, SO_x emissions may be expected to be considerably reduced in a diesel engine using groundnut biodiesel.
- The high flash point (more than 100ºC) of groundnut biodiesel ensures safe storage and safe transportation free from fire hazards. The higher cloud and pour points of groundnut biodiesel than the reference diesel fuel may involve some complications for its use in diesel engine during cold weather.

Groundnut oil and its ethyl ester have good combustion characteristics for use as fuel in a diesel engine. The use of the biodiesel fuel would not constitute a corrosion problem in the injection system and pressure chamber of a diesel engine. Groundnut oil ethyl ester was found to have better fuel quality than raw groundnut oil and it can successfully fuel a diesel engine.

The results of this work contribute to the baseline data needed for future replacement of conventional diesel with renewable biodiesel.

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