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Effect of biodiesel structural configuration on its ignition quality

A. Gopinath[1], Sukumar Puhan[2], G. Nagarajan[3]

[1] Product Development, Ashok Leyland Technical Centre, Chennai, Tamilnadu, India.
[2] Department of Mechanical Engineering, Veltech Engineering College, Chennai, Tamilnadu, India.
[3] Internal Combustion Engineering Division, Department of Mechanical Engineering, College of Engineering, Anna University Chennai, Tamilnadu, India.

Abstract
Biodiesel is an alternative fuel consisting of alkyl esters of fatty acids from vegetable oils or animal fats. The properties of biodiesel depend on the type of vegetable oil used for the transesterification process. Cetane number is one of the most significant properties to specify the ignition quality of any fuel. The cetane number of biodiesel fuels is considerably influenced by their fatty acid methyl ester composition. The objective of this work is to investigate the influence of fatty acid methyl ester composition on cetane number of biodiesel fuels. In the present work, 15 various biodiesel fuels were prepared and their cetane number was measured experimentally. A wide literature review was made and the measured cetane numbers were compared with the reported values. The dependence of cetane number of biodiesel fuels on their fatty acid ester composition investigated. It was found from the investigation that the unsaturation percentage can significantly affect the cetane number.

Keywords: Biodiesel, Cetane number, Fatty acid ester composition, Ignition quality, Unsaturation.

1. Introduction

The diminishing reserves of conventional diesel fuel and indecision in its availability are considered to be the important trigger for many initiatives to look for the alternative source of energy. To reduce the dependence of fossil fuel, research on alternative fuels is being extensively carried out [1, 2]. Organic seed oils such as soybean, sunflower, peanut, cottonseed, rapeseed, coconut, jojoba, jatropha, palm oil and their esters are considered as viable alternative types of fuels because they are renewable, non-toxic and biodegradable and produce lower emissions [3]. The basic composition of any vegetable oil is triglyceride, which is of three fatty acids and one glycerol molecule. A simple molecular representation of a triglyceride is shown in Figure 1.

Biodiesel is obtained when a vegetable oil is chemically reacted with an alcohol to produce mono alkyl esters i.e. biodiesel [4, 5]. In the process, glycerol is obtained as co-product. Biodiesel fuels are generally classified as fatty acid methyl esters (FAMEs) which are derived from transesterification of vegetable oils with methonal, although other alcohols can be used [6]. The fatty acids vary in their carbon chain length and in number of double bond of carbons [7]. Fatty acids may be saturated or unsaturated. A saturated fat is one that cannot chemically accept additional hydrogen and contains only single carbon-

carbon bonds. An unsaturated fat can be hydrogenated and contains one or more double bonds [8]. The structural formula for different fatty acids are listed in Table 1.

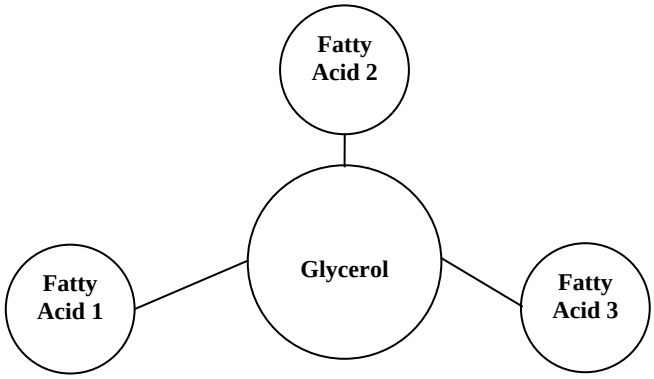

Figure 1. Structure of a triglyceride

Table 1. Structural formula for fatty acids

Acid Chain	C:N	Type	Structure
Caprylic	C8 : 0	S	$CH_3 (CH_2)_6 COOH$
Capric	C10 : 0	S	$CH_3 (CH_2)_8 COOH$
Lauric	C12 : 0	S	$CH_3(CH_2)_{10} COOH$
Myristic	C14 : 0	S	$CH_3(CH_2)_{12} COOH$
Palmitic	C16 : 0	S	$CH_3(CH_2)_{14} COOH$
Palmitoleic	C16 : 1	US	$CH_3(CH_2)_5 CH=CH(CH_2)_7 COOH$
Stearic	C18 : 0	S	$CH_3(CH_2)_{16} COOH$
Oleic	C18 : 1	US	$CH_3(CH_2)_7 CH=CH(CH_2)_7COOH$
Linoleic	C18 : 2	US	$CH_3(CH_2)_4CH=CHCH_2CH=CH(CH_2)_7COOH$
Linolenic	C18 : 3	US	$CH_3CH_2CH=CHCH_2CH=CHCH_2CH=CH (CH_2)_7COOH$
Arachidic	C20 : 0	S	$CH_3(CH_2)_{18}COOH$
Eicosenoic	C20 : 1	US	$CH_3(CH_2)_7CH=CH(CH_2)_9COOH$
Behenic	C22 : 0	S	$CH_3(CH_2)_{20}COOH$
Erucic	C22 : 1	US	$CH_3(CH_2)_7CH=CH(CH_2)_{11}COOH$

where, C:N, indicates C the number of carbons and N the number of double bond of carbons in the fatty acid chain. US = Unsaturated fatty acids and S = Saturated fatty acids

The cetane number is one of the most significant properties to specify the ignition quality of any fuel. The cetane number of esters of vegetable oils (Biodiesel) is greater than those of both vegetable oils and No. 2 diesel fuel [9]. As indicator of ignition quality, the cetane number is a prime indicator of fuel quality in the realm of diesel engines. It is conceptually similar to the octane number used for gasoline. Generally, a compound that has a high octane number tends to have a low cetane number and vice versa. The cetane number of a fuel is related to the ignition delay time, i.e., the time that passes between injection of the fuel into the cylinder and onset of ignition. The shorter the ignition delay time, the higher the cetane number and vice versa. Standards have been established worldwide for cetane number determination, for example ASTM D613 in the United States, internationally the International Organization for Standardization (ISO) standard ISO 5165. A long straight-chain hydrocarbon, hexadecane (C16H34; trivial name cetane, giving the cetane scale its name) is the high quality standard on the cetane scale with an assigned cetane number of 100. A highly branched compound heptamethylnonane (HMN, also C16H34), a compound with poor ignition quality, is the low-quality standard and has an assigned cetane number of 15. The cetane scale is arbitrary and compounds with

cetane number >100 or cetane number <15 have been identified. The accuracy of the definition of heptamethylnonane as a primary reference fuel with a cetane number of 15 has also been called into question. In 1978 Bowden of Southwest Research Institute reported that, using ?-methylnaphthalene and hexadecane as reference fuels, the cetane number of heptamethylnonane was found to be only 12.2, and not 15 as is customarily assigned when it is used as a reference fuel [10].
The standard ASTM D975 for conventional diesel fuel requires a minimum cetane number of 40 while the standards for biodiesel prescribe a minimum of 47 (ASTM D6751) or 51 (EN14214) [11]. The specific objective of the present work is to investigate the effect of biodiesel fatty acid ester composition on its cetane number.

2. Literature review

The influence of fatty acid structure and composition on cetane number was discussed in a number of papers. The cetane number of biodiesel depends on the parent oil source. However, both moieties, the fatty acid chain and alcohol functionality, contribute to the ignition quality of biodiesel fuels [11]. The cetane number of neat fatty compounds decreases with increasing unsaturation and increases with increasing chain length, i.e., uninterrupted CH_2 moities [11]. The branched esters derived from alcohols such as iso-proponal have cetane numbers competitive with methyl or other straight chain alkyl esters [12]. One long chain suffices to impart a high cetane number even if the other moiety is branched. Branched esters are of interest because they exhibit improved low-temperature properties [12]. The cetane number of straight chain fatty acid methyl esters with carbon number of 6, 10, 12, 14, 16, and 18 increased nonlinearly with carbon number [13].
For mono-alkyl esters of fatty acids, long ignition delay times with low cetane numbers and subsequent poorer combustion have been associated with more highly unsaturated components such as the esters of linoleic and linolenic acids [14]. High cetane numbers were observed for esters of saturated fatty acids such as palmitic and stearic acids. The increasing number of double bonds and their positions in their chain causing lower cetane numbers. The more sequential methylene (CH_2) groups in the fatty compound would result in a higher cetane number [14]. The cetane number of saturated fatty acid methyl esters (FAMEs) increases with the longer chain, and the cetane number of unsaturated FAMEs decreases with the degree of unsaturation, or the number of double bonds [15]. According to Michael S. Graboski et. al [8], cetane number increases with chain length, decreases with number of double bonds and decreases as double bonds and carbonyl group move toward the centre of the chain. The authors have also cited that the cetane number of pure esters of stearic acid was approximately 75, but for esters of linolenic acid with three double bonds, cetane number had dropped to the mid-twenties. Cetane number increased from 47.9 to 75.6 for saturated C_{10} through C_{18} esters.At and above C_{12}, the cetane numbers were above 60. G. Knothe [16] has presented that cetane numbers of fatty esters generally increases with :the number of methylene groups (CH_2) in the chain of fatty compound

(i) number of CH_2 groups in the ester moiety
(ii) with increasing saturation of the fatty compound.

In addition G. Knothe observed that the cetane number also depends on the position of the double bond in monounsaturated fatty compounds and increases if the double bond move towards one end of the molecule.
Yamane et al.[17] investigated the effect of fatty acid methyl esters in fuels on the ignition delay of diesel combustion using oleic acid methyl ester and linoleic acid methyl ester.They have shown that the cetane number of oleic acid methyl ester is higher than that of linoleic acid methyl ester, resulting in shorter ignition delay of oleic acid methyl ester compared with linoleic acid methyl ester. E. Klopfenstein [18] has reported the cetane numbers of series of methyl esters of the saturated fatty acid esters from 8 to 18 carbons and the effect of fatty acid chain length on cetane numbers. The author has mentioned that the cetane number increases in a nonlinear manner with increase in chain length of fatty acid for methyl esters. It was also reported that the cetane number increases with increase in molecular weight for esters of the normal alcohols when fatty acid was kept constant. The cetane number increase resulting from an increase in the molecular weight of the alcohol of the ester was less than that for the same increase in the molecular weight of the fatty acid portion of the ester. The data points and trend curve of cetane number of methyl esters are shown in Figure 2. The data points were taken from [18] and [19].
The overall biodiesel properties including cetane number are strongly influenced by the properties of individual fatty acid esters in biodiesel. It therefore looks sensible to supplement particular fatty ester(s) with desirable properties in the fuel to upgrade the properties of whole fuel. It may be attainable in the

future to improve the properties of biodiesel by means of genetic engineering of the parent vegetable oils, which could ultimately lead to a fuel enriched with certain fatty acids, those exhibit a combination of improved fuel properties [11].

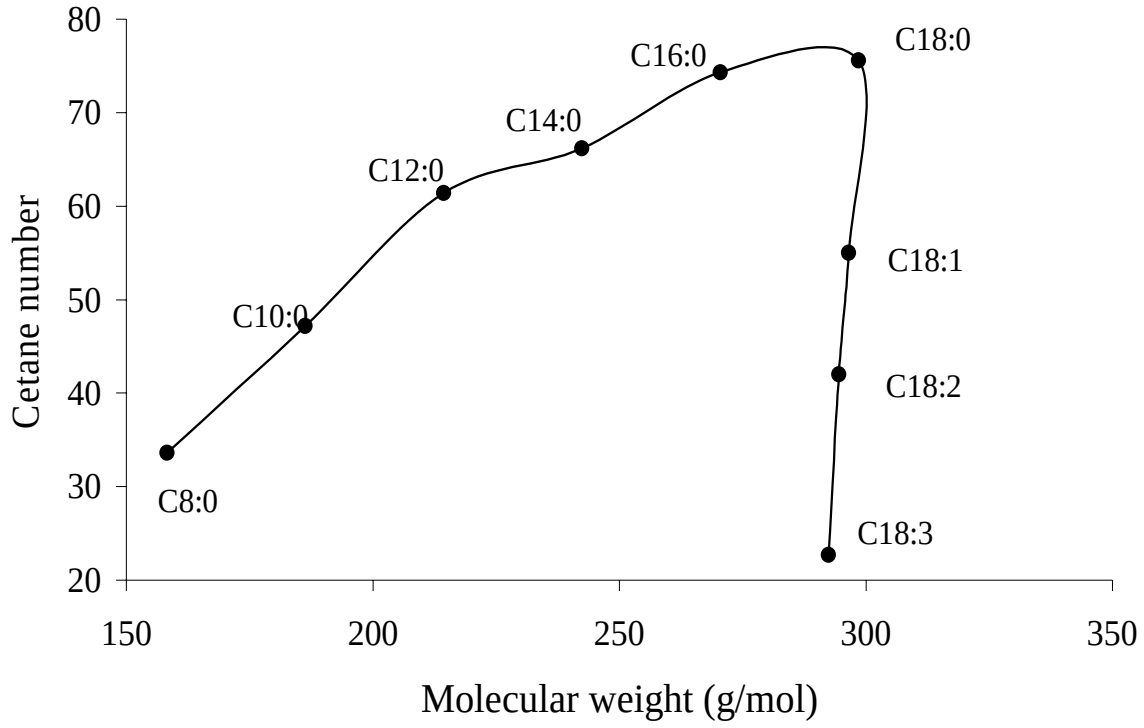

Figure 2. Cetane number trend lines for methyl esters

3. Present work

In the present work, 15 different biodiesel (including three blends) fuels were prepared and their properties were experimentally measured following the methods specified in the ASTM standards. The measured cetane numbers were compared with literature values and the difference were reported. The measured cetane numbers show a good agreement with the literature values. The effect of fatty acid methyl ester composition on cetane number was investigated.

4. Experiments

4.1 Biodiesel preparation

The raw oils (Rapeseed, Soybean, Rubberseed, Cottonseed, Jatropha, Karanja, Neem, Sunflower, Palm, Tallow, Coconut and Mahua) were purchased from Annai Biocrop Pvt. Ltd, Chennai. The biodiesels were produced in the authors' laboratory through transesterification. Transesterification is a process of producing a reaction in triglyceride and alcohol in the presence of a catalyst to produce alkyl ester and glycerol. Alkali (Sodium hydroxide, pottosium hydroxide), acids (Sulphuric acid Hydro chloric acid) catalyze reaction [20, 21, 22]. Alkali catalyzed transesterification is faster than acid catalyzed transesterification and is most used commercially [23]. If the free fatty acid (FFA) content and moisture content are less than 0.5 %, good quality of biodiesel can be produced. The objective of the transesterification process is to reduce the viscosity of vegetable oil. In the present work, biodiesel fuels were produced by single stage transesterification process. The two stage and three stage transesterification processes are not discussed since it may not required for the present discussion. The selection of transesterification process and step-by-step procedures to produce biodiesel fuels are mentioned in Appendix 1 and 2.

4.2 Fuel properties

The fuel properties were determined following the methods specified in ASTM standards as given in Table 2 for different biodiesel fuels. The fatty acid composition and free fatty acid content were measured in Tamil Nadu Oil Seeds Association, Chennai. The cetane number and moisture content were

measure in ITALAB Pvt. Ltd, Industrial Testing and Analytical Laboratories (An ISO 9001 : 2000 Certified Organization), Chennai.

Table 2. ASTM methods for determination of fuel properties

Property	Unit	Standard
Fatty Acid Composition	wt %	D 6584
Cetane Number	-	D 613
Moisture Content	% volume	D 2709

5. Results and discussion

The results of fuel tests on different biodiesel fuels are summarized in Table 3 and 4. The measured cetane numbers were compared with the available published data and listed in Table 5.

However, except for rapeseed and sunflower biodiesel fuels, the measured cetane numbers show a good agreement with the published data.

A correlation analysis was made to find out the degree of linear association between the cetane number and different fatty acids. The percentage of unsaturation was also considered in this analysis. The Pearson product moment correlation coefficient between different properties and percentage of unsaturation is shown in Table 6. The formula used to find out the Pearson correlation coefficient (r) is shown in equation (1).

$$r = \frac{\left(X - \bar{X}\right)\left(Y - \bar{Y}\right)}{\sqrt{\left(X - \bar{X}\right)^2\left(Y - \bar{Y}\right)^2}} \quad (1)$$

Table 3. Fatty acid composition of different biodiesel fuels and blends

Biodiesel	Fatty Acid Methyl Ester Composition (wt %)								% of US	% of S
	Lauric	Myristic	Palmitic	Stearic	Oleic	Linoleic	Linolenic	Others		
Rapeseed	0.0	1.0	3.5	0.9	64.1	22.5	8.0	0.0	94.6	5.4
Soybean	0.1	0.1	10.2	3.7	22.8	53.7	8.6	0.8	85.1	14.1
Rubber seed	0.0	0.2	12.5	8.3	27.8	37.7	13.4	0.1	78.9	21.0
Cottonseed	0.1	1.0	20.1	2.6	19.2	55.2	0.6	1.2	75.0	23.8
Jatropha	0.0	0.1	15.6	10.5	42.1	30.9	0.2	0.6	73.2	26.2
Karanja	0.0	0.1	9.9	7.8	53.2	19.1	0.0	9.9	72.3	17.8
JP 50:50	0.1	0.5	28.1	7.7	42.7	20.1	0.2	0.6	63.0	36.4
Neem	0.8	0.5	18.2	20.1	41.3	16.4	0.3	2.4	58.0	39.6
Sunflower	0.2	0.8	38.6	4.6	44.0	10.7	0.1	1.0	54.8	44.2
Palm	0.2	0.8	39.5	5.1	43.1	10.4	0.1	0.8	53.6	45.6
Mahua	0.0	0.2	20.8	25.2	36.4	15.8	0.3	1.3	52.5	46.2
SFCt 50:50	20.3	10.5	9.3	4.3	19.4	32.6	0.0	3.6	52.0	44.4
Beef Tallow	0.1	2.5	23.3	19.4	42.4	2.9	0.9	8.5	46.2	45.3
JCt 50:50	20.9	10.4	13.7	7.2	26.1	18.2	0.1	3.4	44.4	52.2
Coconut	45.6	22.1	10.2	3.6	8.2	2.7	0.0	7.6	10.9	81.5

where,
JP 50:50 = Blend of 50 % Jatropha and 50 % Palm by Volume,
SFCt 50:50 = Blend of 50 % Sunflower and 50 % Coconut by Volume,
JCt 50:50 = Blend of 50 % Jatropha and 50 % Coconut by Volume,
% US = % of Unsaturated Fatty Acids, and
% S = % of Saturated Fatty Acids in biodiesel.

Table 4. Measured properties of different biodiesel fuels and blends

Biodiesel	Cetane Number	Moisture content (wt %)	Free Fatty Acid (FFA) content
Rapeseed	46.0	0.02	0.08
Soybean	48.0	0.05	0.05
Rubber seed	51.0	0.03	0.11
Cottonseed	52.1	0.02	0.09
Jatropha	54.0	0.03	0.03
Karanja	52.0	0.02	1.09
JP 50:50	59.0	0.03	0.44
Neem	58.7	0.06	0.03
Sunflower	61.6	0.03	0.12
Palm	64.0	0.02	0.46
Mahua	61.4	0.08	0.02
SFCt 50:50	54.6	0.05	1.04
Beef Tallow	58.8	0.04	0.11
JCt 50:50	58.0	0.06	1.06
Coconut	60.0	0.07	0.08

Table 5. Comparison of measured cetane numbers with published data

Biodiesel	Cetane Number			
	Measured value	Literature values	Average of Literature values	% Difference
Rapeseed	46.0	52.9 [8], 54 [15], 51[24], 51[25]	52.2	-12
Soybean	48.0	50.9 [8], 45 [26], 45 [27], 51.61[28], 51.5 [29]	48.8	-2
Rubber seed	51.0	NA	-	-
Cottonseed	52.1	51.2 [8], 52 [30]	51.6	1
Jatropha	54.0	50 [31], 52.31 [32], 58.4 [33]	53.6	1
Karanja	52.0	53.9 [33], 51[34]	52.5	-1
JP 50:50	59.0	NA	-	-
Neem	58.7	NA	-	-
Sunflower	61.6	61.2 [25], 49 [26], 49 [27], 50[30], 58 [35]	53.4	15
Palm	64.0	64.5 [15], 53 [24], 70 [25], 62 [26], 62 [27],	62.3	3
Mahua	61.4	65.4 [33]	65.4	-6
SFCt 50:50	54.6	NA	-	-
Beef Tallow	58.8	58.8 [8], 57.78 [28], 52 [36],	56.2	5
JCt 50:50	58.0	NA	-	-
Coconut	60.0	57 [15]	57.0	5

where, NA = Not Available, The number within the square brackets indicates the literature reference number. and % Difference = % Difference between measured value and average of Literature values

From Table 6, it can be found that the the methyl ester of lauric, myristic, palmitic, and stearic fatty acids are positively correlated with cetane number. On the other hand, the methyl ester of oleic, linoleic, and linolenic fatty acids are negatively correlated with the cetane number. However, the correlation coefficients for lauric, myristic, and oleic are not so significant, the correlation analysis was done only to realize the trend between these fatty acids and cetane number. The cetane number of pure fatty acid methyl esters are listed in Table 7. Before getting into to the investigation findings, it is necessary to have a look at the cetane number of individual fatty acids and their methyl esters.

Table 6. Pearson correlation coefficient (r) between cetane number and fatty acid methyl esters

"X" variable	"Y" variable	Correlation Coefficient
Lauric	Cetane number	0.210
Myristic		0.221
Palmitic		0.746
Stearic		0.415
Oleic		-0.093
Linoleic		-0.699
Linolenic		-0.671
% of US		– 0.760

Table 7. Cetane number of pure fatty acid methyl esters

Fatty Acid	Fatty Acid Methyl Ester	Molecular weight of FAMEs (g/mol)	Cetane Number of FAMEs	Average of cetane no.
Lauric	Lauric acid methyl ester (Methyl Laurate)	214.35 [4]	60.8 [13], 61.4 [15]	61.1
Myristic	Myristic acid methyl ester (Methyl Myristate)	242.40 [4]	73.5 [13], 66.2 [15]	69.9
Palmitic	Palmitic acid methyl ester (Methyl Palmitate)	270.46 [4]	74.3 [13], 85.9 [14], 74.5 [15], 74.3 [19], 74 [37],	76.6
Stearic	Stearic acid methyl ester (Methyl Stearate)	298.51 [4]	101[11], 75.6 [13], 101.0 [14], 86.9 [15], 75.6 [19], 75 [37]	85.9
Oleic	Oleic acid methyl ester (Methyl Oleate)	296.49 [4]	59.3 [11], 59.3 [14], 56.0 [15], 55 [19], 55 [37]	56.9
Linoleic	Linoleic acid methyl ester (Methyl Linoleate)	294.48 [4]	38.2, and 42.2[11], 38.2 [14], 41.7 [15], 42 [19], 33 [37],	39.2
Linolenic	Linolenic acid methyl ester (Methyl Linolenate)	292.46 [4]	20.6 and 22.7 [11], 45.9 [15], 22.7 [19]	28.0

From Table 7, it can be noticed that the methyl ester of stearic acid has a higher cetane number than that of other FAMEs. This is due to the fact that methyl stearate has a higher molecular weight than that of other FAMEs. Methyl linolenate on the other hand has a lower cetane number than the other FAMEs. This is because cetane number decreases with number of double bonds or unsaturation. The relationship between fatty acid methyl ester composition and the cetane number of biodiesel fuels were investigated.

FromTable 3 and 4 it can be observed that rapeseed is highly unsaturated (around 94 %) than other biodiesel fuels. The cetane number of rapeseed is lower than that of other biodiesel fuels. Although coconut oil is more saturated (around 81 %) than other biodiesel fuels, it has lower cetane number than palm, sunflower and mahua. It is due to the contribution of palmitic acid (palm 39.51 %, sunflower 38.63 %, mahua 20.84 % and coconut oil 10.21 %) and stearic acid (palm 5.09 %, sunflower 4.55 %, mahua 25.15 % and coconut oil 3.57 %). From the discussions it may be concluded that the overall percentage of unsaturation or saturation may not be sole authority to decide cetane number of a biodiesel. The contribution of long straight chain fatty acids can have a significant influence on the cetane number of biodiesel.

From Table 4, the cetane number of jatropha and palm biodiesels are 54 and 64 respectively. Whereas the cetane number 50:50 blend of jatropha:palm biodiesel is 59. This may direct to an idea that the cetane number of 50:50 blend fuels is an average of the cetane number of the two fuels. This is not true for other

blends. Because the cetane number of 50:50 blend of sunflower:coconut biodiesel is 54.6 which is not an average of cetane numbers of sunflower (cetane number = 61.6) and coconut oil biodiesel fuels.(cetane number = 60). Similarly, the the cetane number of 50:50 blend of jatropha:coconut is 58 which is not equal to the average cetane number of these two biodiesel fuels. From Table 8, the correlation coefficient between cetane number and percentage of unsaturation can be observed as (–) 0.760. Figure 3 illustrates the variation of cetane number with percentage of unsaturation. The figure shows a declining trend between cetane number and percentage of unsaturation.

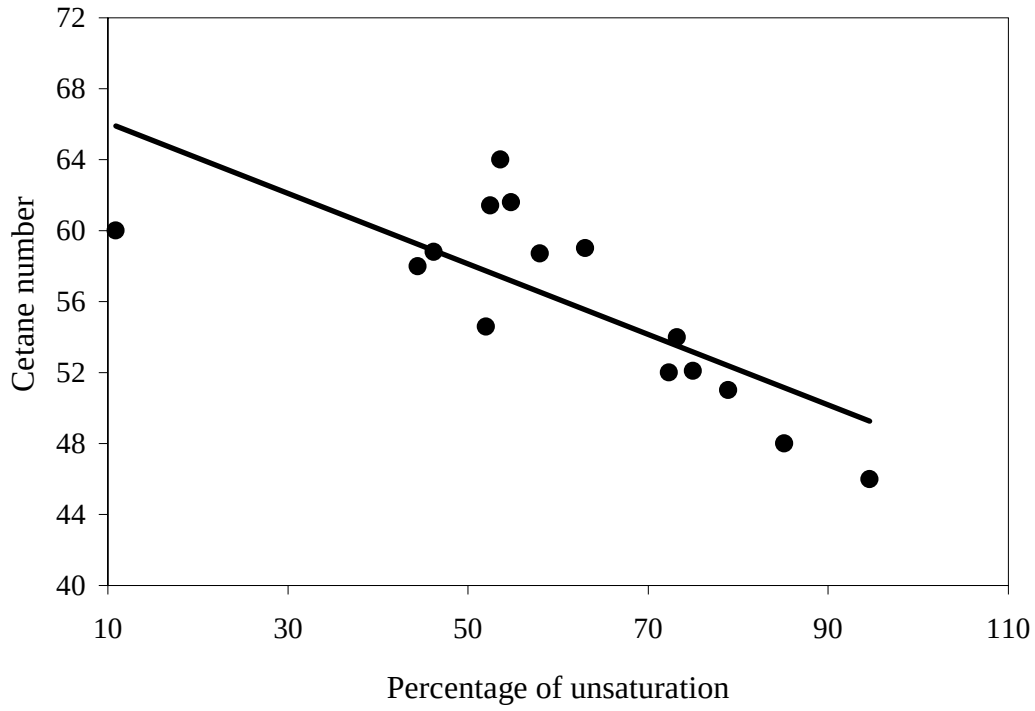

Figure 3. Variation of cetane number with percentage of unsaturation

6. Conclusions

From the present work, it can be concluded that the cetane number of biodiesel fuels is strongly influenced by the individual fatty esters in the biodiesel. The increasing number of double bonds causing lower cetane numbers. Cetane number increases with increasing chain length and decreases with increasing unsaturation. The cetane number increases with increase in lauric, myristic, palmitic, and stearic fatty acids content. Whereas cetane number decreases with increase in olelic, linoleic, and linolenic fatty acid ester content.

Appendix 1: Selection of transesterification process

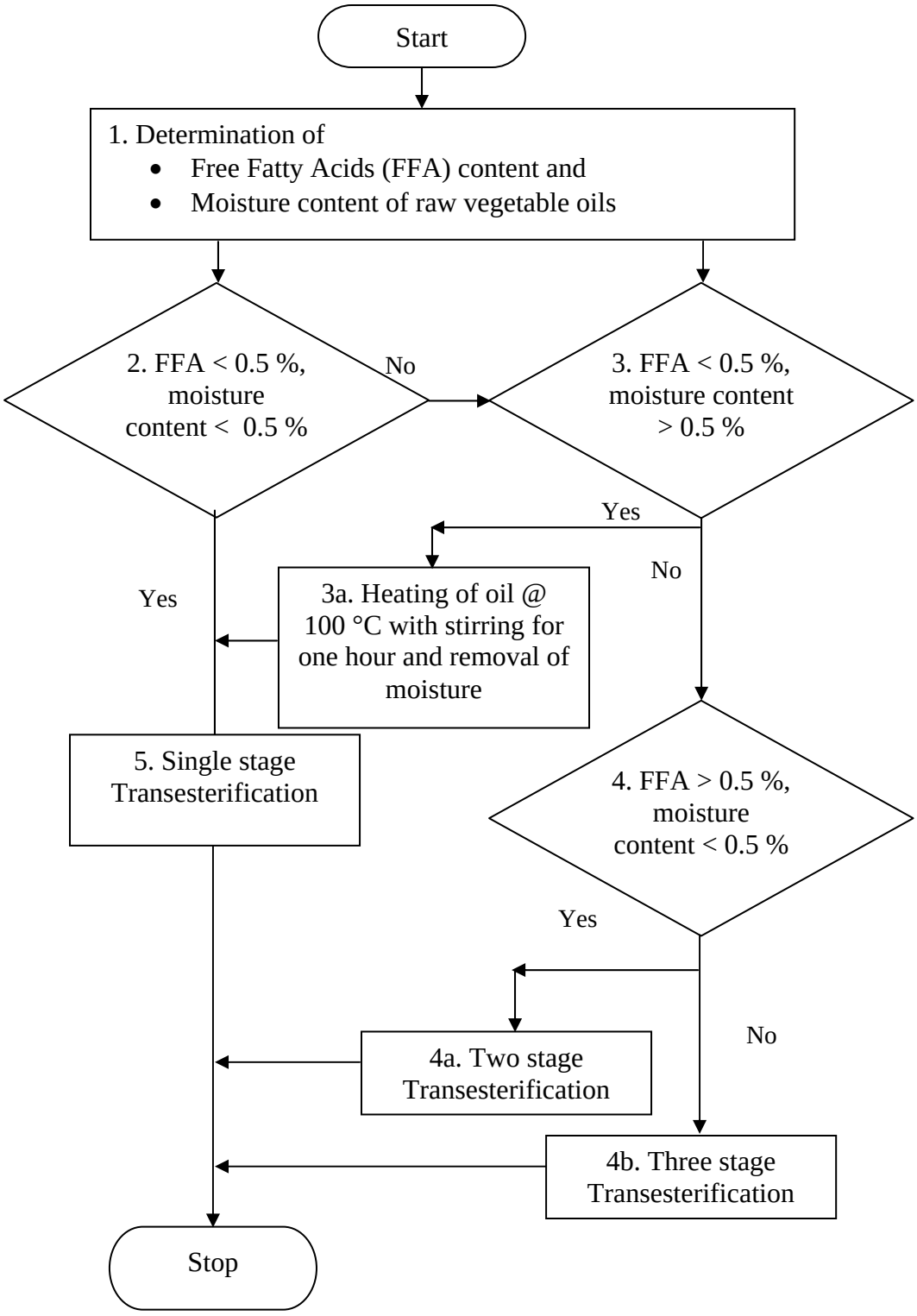

Figure A1. Flow chart for the selection of transesterification process

Appendix 2: Step-by-step procedure to produce biodiesel fuels

1. The parent vegetable oil was poured in a three necked flask.
2. The oil was heated using heating mantle.
3. In Parallel, methoxide (methanol + sodium hydroxide (NaOH) or methanol + potassium hydroxide (KOH), in the present work, NaOH used as catalyst) was prepared. The mixing ratio was taken as 1 kg oil: 200 g of methanol: 5 g of NaOH (generally the alcohol and the catalyst quantity are 20 % and 0.3 to 0.5 % of oil respectively). The mixture of methanol and NaOH stirred manually till all the NaOH diffused in the methanol.
4. The prepared methoxide was then poured in the three necked flask that contains the heated oil when it reached to 50°C. The whole mixure in the flask was continuosly stirred by a magnetic stirrer.The speed of the stirred was maintained at about 250 m/s (speed can be varied according to the mixture quantity).
5. One neck of the flask closed using thermowell and a thermometer inserted along with the thermowell. The second neck closed with a stopper to avoid the contact between system and atmosphere. The last neck closed with a condenser. The cooling water temperature maintained at dew point temperature of alcohol (about 20°C).
6. Every 10 minutes, the temperature of the reactant was measured with the help of thermometer. The regulator in the heating mantle was adjusted to maintain the reactant temperature at 60°C (the boiling point of methanol is 60°C).
7. The reaction process was allowed for about two hours. After two hours, the heating mantle was switched off to stop the reaction process.
8. All the three necks of the flask opened and the mixture was transferred from the flask to a two litre separating funnel.
9. The mixture was allowed in the funnel for about eight hours. The mixture was separated into two layers as mono alkyl ester (biodiesel) and glycerol.
10. Due to the higher density of glycerol, it settled at the bottom.
11. The glycerol alone was removed out through the opening of the funnel.
12. To remove the alcohol and catalyst present in the ester, the ester was washed with distilled water.
13. 120 g (generally 10 % of ester by mass) of distilled warm water at 40°C (generally at 35 – 40°C) poured in the ester.
14. The ester and the water were allowed in the funnel to eight hours. The water (milky white colour) was settled at the bottom due to higher density.
15. The water was removed out through the opening of the funnel.
16. The ester (biodiesel) was heated again and maintained at 100°C about an hour to remove the moisture content. And thus the required biodiesel was made ready.

Following the procedures mentioned above, all the other biodiesel fuels were produced. By mixing 50 % by volume of two different biodiesel fuels, the relevant blends were prepared.

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A. Gopinath has completed master of engineering in internal combustion engineering with gold medal from College of Engineering, Guindy, Anna University Chennai, Chennai, Tamilnadu, India in 2008. He is currently working as Deputy Manager- Product development in Ashok Leyland Technical Centre, Chennai, India.
His area of research includes biodiesel composition, properties, their optimization and utilization of biodiesel in diesel engines.
E-mail address: gopinathmeice@yahoo.co.in, Mob: +91 9840175008.

Sukumar Puhan has done his doctorate in internal combustion engineering discipline in the area of biodiesel from Anna University Chennai, Tamilnadu, India in 2009. He is currently working as a Professor in Department of Mechanical Engineering, Veltech Engineering College, Chennai, India. He has published a significant number of international articles.
His research areas include biodiesel cultivation to its usage in diesel engines, biodiesel genetic modification, and analysis of the effect of biodiesel composition in diesel engine's combustion, performance, and emission characteristics.
E-mail address: sp_anna2006@yahoo.co.in, Mob: +91-9444489013.

G. Nagarajan has done his doctorate in internal combustion engineering discipline from Anna University Chennai, Tamilnadu, India in 2000. He is currently working as a Professor in Department of Mechanical Engineering, College of Engineering Guindy, Anna University, Chennai, India. He is also the Director of Entrance Examinations, Anna University, Chennai. He has published more than 40 international articles, 37 international conference papers, 9 national journal papers, and 21 national conference papers. He is currently handling 4 consultant projects and 3 testing projects.
E-mail address: nagarajan1963@annauniv.edu, Mob:+91 9445393075.

