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Prediction of cetane number of biodiesel fuel from the fatty acid methyl ester (FAME) composition

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A b s t r a c t. Cetane number is an important parameter in evaluating the quality of biodiesel fuel. Its determination is usually arduous and expensive, and the results obtained are not always accurate due to experimental error. This work is aimed at developing a relationship between the fatty acid methyl ester (FAME) composition and the cetane number (CN).

Data were collected from the literature on the cetane number of various biodiesel fuels and their FAME composition. A regression analysis was performed on pure FAME to establish a relationship between the cetane number and FAME composition.

An equation was developed relating the cetane number with the % composition by weight of FAME of biodiesel fuel. The predicted cetane number values compare well with the average measured cetane number values. This work establishes the general dependence of cetane number on FAME composition of biodiesels.

K e y w o r d s: cetane number, biodiesel, FAME composition, regression analysis, prediction

INTRODUCTION

Emissions of hazardous gases from the exhausts of heavy duty vehicles have increased tremendously over the years. This has resulted in intense air pollution, identified to be one of the reasons for climatic changes that result in frequent heavy rains, hurricanes and floods threatening lives and properties. There is a need to reduce carbon-dioxide and other hazardous emissions to the atmosphere in internal combustion engines to decrease air pollution. Added to this is the quest to reduce dependence on petroleum products as a result of the fast depletion of crude oil resources and instability in its price. This has shifted the focus on sourcing for an alternative energy from other materials than petroleum products.

Pure vegetable oils have been considered as alternatives for diesel fuel, but the high viscosity at room temperature made them unsuitable for diesel engines. However, fatty acid methyl ester (FAME) have lower viscosity than the pure oils, and as such are promising alternatives. While the focus has been mostly on oils like soybean, rapeseed, and sunflower (Lang *et al.*, 2001), which are essentially edible in nature, biodiesel has also been produced from non-edible sources such as used frying oil and greases (Alcantara *et al*., 2000; Canakci and Gerpen, 2001; Mittelbach *et al*., 1992).

Attempts have been made by various researchers to determine the best composition of biodiesel that would enhance the combustion process. It was observed that the fuel properties of biodiesel play a significant role in the combustion process. One of such properties is the influence of the Cetane Number (CN) on the combustion process and on engine performance. The CN is a commonly used indicator for the determination of diesel fuel quality, especially the ignition quality. It measures the readiness of the fuel to auto-ignite when injected into the engine. Ignition quality is one of the properties of biodiesel that is determined by the structure of the FAME component (Knothe, 2005).

The nature of the fuel components determines the fuel properties. One of those in this case is the structure of the FAME comprised in biodiesel. The CN of biodiesel depends on the distribution of fatty acids in the original oil or fat from which it was produced. The major factor affecting FAME profiles is the plant itself and variation will occur, depending on climate and processing methods (Michael and Robert, 1998; Fangrui and Milford, 1999). However, under proper conditions, the fatty acid profile should remain largely unchanged during processing. Van Gerpen (1996) in his

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work on CN testing of biodiesels observed that CN of various level of FAME composition of soybean depends on the distribution of fatty acids in the original oil or fat. The longer the fatty acid carbon chains and the more saturated the molecules, the higher the CN (Geller and Goodrum, 2004). A high value of CN was observed in saturated FAME such as palmitate and stearate, while the CN is in the medium range in mono-unsaturated FAME. This is due to increasing number of double bonds, and the branching in the chain, causing lower CN values. The CN decreases with increasing unsaturation and increases with increasing chain length.

A wide variation in the values of the CN for biodiesel has been reported (Table 1). The values of CN of soybean oil-derived biodiesel ranged from 45 to 60, while those of rapeseed oil-derived biofuel ranged from 48 to 61.2. The CN value varies from feedstock to feedstock, depending generally on the chemical composition of the fuel, and can affect the engine ability to start, the noise level, and the exhaust emissions. It has been observed that feedstock of the same type of oil do vary in CN, though much less than the difference between oil sources (Van Gerpen, 1996). A considerable variation in the CN values from different laboratories was observed, and even between duplicate samples of the same laboratory.

The test procedure of CN has been subjected to criticism based on the differences between the cetane test engine configuration and operating conditions, and the configurations and operating conditions of modern engines. The CN test procedure is complex and inherently involves considerable uncertainty due to experimental error. It is not always easy conducting engine tests to determine CN due to the cost of the reference fuels and the effort required.

Cetane number is measured using blends of two reference fuels, namely *n*-cetane (100 CN) and hepta methylnonane (15 CN). Cetane number of the test fuel is the percentage by volume of *n*-cetane in a blend of *n*-cetane (100 CN) and hepta methylnonane (15 CN) having the same ignition quality when tested in the same engine under the same test conditions (ASTM, 2004).

One of the ways of solving the problem of CN determination is to develop models to predict the CN when some parameters are known. Since it has been established that the composition of FAME affects CN, models can be developed relating CN to the composition of FAME. Ramadhas *et al.* (2006) used artificial neural network (ANN) models to predict the CN of some fuels based on their FAME composition. The FAME composition of biodiesel and the experimental CN values were used to train the networks. The fatty acid composition, in percentage of various esters in pure form as well as mixtures of esters (biodiesel) and their experimental CN, are taken into account for developing the present model (Ramadhas *et al*., 2006). A computer code was developed in MATLAB to implement the present ANN models. Four types of networks have been chosen to predict

T a b l e 1. Reported values of the cetane number for biodiesel

SME*	RME	SUNME	CME	PME	POME	LME	TME	CAME
45.0 [1]**	51.9 [14]	61.2 [19]	52.0 [9]	54.0 [4]	59-70 [28]	63.6 [10]	58.0 [37]	53.9 [13]
46.2 [2]	54.4 [15]	50.0 [9]	45-55 [25]	54.0 [8]	62.0 [8]	-	62.9 [13]	55.0 [13]
54.7 [3]	49.9 [16]	58.0 [23]	-	-	62.4 [26]	-	64.8 [13]	-
45.0 [4]	54.5 [17]	58.0 [24]	-	-	58.3 [27]	-	-	-
54.8 [5]	54-65 [18]	49.0 [8]	-	-	62.0 [28]	-	-	-
60.0 [6]	53.0 [9]	-	-	-	-	-	-	-
51.9 [7]	51-59 [19]	-	-	-	-	-	-	-
48.6 [7]	51.0 [20]	-	-	-	-	-	-	-
45.0 [8]	59.7 [21]	-	-	-	-	-	-	-
51.0 [9]	44-48 [22]	-	-	-	-	-	-	-
47.2 [10]	53.3 [28]	-	-	-	-	-	-	-
52.0 [11]	-	-	-	-	-	-	-	-
56.4 [12]	-	-	-	-	-	-	-	-
47.3 [13]	-	-	-	-	-	-	-	-
59.0 [13]	-	-	-	-	-	-	-	-

*SME – soybean methyl ester, RME – rapeseed methyl ester, SUNME – sunflower methyl ester, CME – cottonseed methyl ester, PME – peanut methyl ester, POME – palm oil methyl ester, LME – lard methyl ester, TME – tallow methyl ester, CAME – canola methyl ester.
**The numbers in the parenthesis represent the different authors from which the cetane numbers were sourced from literatures for this work: 1. Reed, 1993; 2. Wagner *et al*., 1984; 3. McDonald *et al.,* 1995; 4.Pischinger *et al.,* 1982; 5. Peterson *et al.,* 1994; 6. Sharp, 1994; 7. Midwest Biofuels, 1993; 8. Marchetti *et al*., (in press); 9. Rakopoulos *et al.,* 2006; 10. McCormick, 2001; 11. Van Gerpen *et al.,* 2004; 12. Wang *et al.*, 2000; 13. Kinast, 2003; 14. Gragg, 1994; 15. Peterson *et al.,* 1993; 16. Mittlebach *et al.,* 1985; 17. Reece and Peterson, 1993; 18. Vellguth, 1983; 19. Takesawa, 1993; 20. Labeckas and Slavinskas, 2006a; 21. Khan, 2002; 22. Labeckas and Slavinskas, 2006b, 23. Arkoudeas *et al.,* 2003; 24. Kalligeros *et. al*., 2003; 25. Altin *et al*., 2001; 26. Liang *et al*., 2006; 27. Schwab *et al.*, 2000, and 28. Knothe, 2005.

the CN of biodiesel. These are the multi-layer feed forward (MLFFN), radial base function (RBFN), generalized regression (GRNN) and recurrent network (RNN). The predicted CN of biodiesel is comparable to that of actual CN of the biodiesel, and it has been concluded that the CN of biodiesel can be predicted based on FAME composition. However, there is no empirical equation for predicting the CN. There is, therefore, a need for an empirical equation for estimating the CN.

The focus of this work was to develop the regression equations for predicting the CN of biodiesel based on the FAME chain compositions, and to verify the applicability of the developed equations for predicting the CN based on the FAME composition of the vegetable oil used for biodiesel production.

MATERIALS AND METHODS

Selection of fuel data

FAME compositions of vegetable oil and CN of pure methyl esters were obtained from different reports from testing laboratories using ASTM D 613 as shown in Tables 2 and 3. Average values of the data collected for CN of pure methyl esters (Table 3) were used in this work.

Regression analysis

A nine by eight matrix was formed with CN as the dependent variable and the pure FAME composition as the independent variables as shown in Table 4. A linear regression analysis was performed on the average values of cetane number data of the pure FAME (Tables 2 and 4).

The regression equation used was as follows:

$$CN = K + ax_1 + bx_2 + cx_3 + dx_4 + ex_5 + fx_6 + gx_7 + hx_8 , \quad (1)$$

where: CN – cetane number; K, a, b, c, d, e, f, g, h are constants to be determined in the regression analysis; $x_1 \ldots\ldots x_8$ are % compositions of FAME.

RESULTS AND DISCUSSION

Substituting the values of K, a – h in Eq. (1) for the coefficients obtained during regression analysis we obtain:

$$CN = 61.1 + 0.088x_2 + 0.133x_3 + 0.152x_4 - 0.101x_5 - 0.039x_6 - 0.243x_7 - 0.395x_8 . \quad (2)$$

Equation (2) shows the relationship between the CN and the FAME composition. From the equation, it is observed that the coefficient for the saturated FAME is positive and increases with an increase in the carbon number from C12:0 to C16:0. This suggests an increase in the CN number with an increase in the composition of the saturated FAME. However, the coefficient of the unsaturated FAME is negative, showing a reduction in the overall CN with unsaturation and further reduces with increase in the number of carbon and unsaturation. This equation is in agreement with the findings of other authors that saturated compounds have higher CN than the unsaturated compounds (Gragg, 1994; Van Gerpen, 1996). The same trend is observed in Table 2 for pure methyl esters. This indicates that FAME composition will play a dominant role in establishing the CN.

The results of the prediction of CN values on the basis of the test data as shown in Fig. 3 indicated that the equation developed was able to predict the CN accurately. The equation developed was applied to other results obtained by different researchers. The results obtained gave an indication of variation of the CN depending on the % composition

T a b l e 2. Cetane number for pure FAME

Fuel	Cetane number	Reference
Caprylic acid methyl ester	18.0	Freedman and Bagby, 1990
	33.6	Knothe, 2005
	47.9	Freedman and Bagby, 1990
	47.2	Knothe, 2005
Lauric acid methyl ester	60.8	Freedman and Bagby, 1990
	61.4	Knothe, 2005
	61.4	McCormick *et al*., 2001
Myristic acid methyl ester	73.5	Freedman and Bagby, 1990
	66.2	Knothe, 2005
Palmitic acid methyl ester	74.3	Freedman and Bagby, 1990
	74.5	Knothe, 2005
Stearic acid methyl ester	75.6	Freedman and Bagby, 1990
	86.9	Knothe, 2005
Palmitoleic acid methyl ester	51.0	McCormick *et al*., 2001
Oleic acid methyl ester	55.0	Knothe *et al*., 1997
	59.3	McCormick *et al*., 2001
	56.0	McCormick *et al*., 2001
Linoleic acid methyl ester	41.7	McCormick *et al*., 2001
	42.2	Knothe *et al*., 1997
	38.0	McCormick *et al*., 2001
	22.7	Knothe *et al*., 1997
	20.4	Knothe *et al*., 1997
	45.9	McCormick *et al*., 2001

T a b l e 3. Fatty acid compositions of methyl esters of different feedstocks

Ref.**	Lauric	Myristic	Palmitic	Stearic	Palmitoleic	Oleic	Linoleic	Linolenic
SME*								
[1]	0.1	0.1	10.2	3.7	0	22.8	53.7	8.6
[2]	0	0	12.9	3.7	0.1	22.2	52.9	7.9
[3]	0	0	10.76	4.37	0	24.13	51.83	6.81
[4]	0	0	10.5	3.9	0	23.3	53	7.6
[5]	0	0.56	14.17	5.19	1.27	48.2	22.19	1.45
[6]	0	0	10.7	3.2	0	25	53.3	5.4
[7]	0	0	3.86	3.9	0	77.9	13.31	0
[8]	0	0	11.75	3.15	0	23.26	55.53	6.31
[9]	0	0	10.4	4.7	0.35	24.8	52.5	6.5
[10]	0	0.1	10.3	4.7	0	22.6	54.1	0.6
[11]	0	0.9	10.54	3.75	1.3	23.18	48.92	1.16
CAME								
[12]	0	0	6.8	3.26	0	16.93	73.73	0
SUME								
[8]	0	0	5.79	5.68	0	20.43	66.02	0
[4]	0	0	6.1	4.1	0	18.6	69	0.3
[9]	0	0	6.06	4.8	0	20.5	67.7	0.4
[12]	0	0	6.38	4.09	0	23.68	63.29	0.36
[16]	0	0	8.6	1.93	0	11.58	77.89	0
[6]	0	0	6.1	4.2	0	24	63.5	0.4
POME								
[16]	0.1	1	42.8	4.5	0	40.5	10.1	0.2
[13]	0	0	42.6	4.4	0.3	40.5	10.1	0.2
[4]	0.3	1.1	42.7	4.3	0	40.9	9.2	0.2
[17]	0.26	1.09	44.81	4.09	0.2	39.99	8.94	0.27
[17]	0.38	0.98	43.32	3.81	0.2	40.57	10.25	0.25
[9]	0.2	1.11	40.2	4.5	0.42	43.3	9	1
[14]	0.46	1.22	47.9	4.23	0.04	37	9.07	0.26
[10]	0.9	1.3	43.9	4.9	0	39	9.5	0.3
[6]	0	0	39.5	4.1	0	43.2	10.6	0.2
TME								
[16]	0.1	2.8	23.3	19.4	0	42.4	2.9	0.9
[10]	0.1	0.1	25.2	19.2	0	48.9	2.7	0.5
[4]	0.1	2.3	23.1	15.4	0	45.8	3.5	0.6
[4]	0	0	23.9	17.5	0	43.9	2.3	0.1
[11]	0.6	2.91	24.34	19.1	3.44	40.23	2.58	3.3
CME								
[16]	0.1	0.7	20.1	2.6	0	19.2	55.2	0.6
[13]	0	0	28.7	0.9	0	13	57.4	0
[8]	0	0	11.67	0.89	0	13.27	57.51	0
[16]	0	0	28.33	0.9	0	13.27	57.51	0
[9]	0	1.2	20.6	4.6	0.8	19.54	52.5	0.4
[10]	0	0	22.9	3.1	0	18.5	54.2	0.5

T a b l e 3. Continuation

Ref.*	Lauric	Myristic	Palmitic	Stearic	Palmitoleic	Oleic	Linoleic	Linolenic
PME								
[16]	0	0	11.38	2.39	0	48.28	31.95	0.93
[13]	0	0	11.4	2.4	0	48.3	32	0.9
[4]	0	0	11.2	3.2	0	45.5	31.7	0.4
[10]	0	0	10.4	8.9	0	47.1	32.9	0.5
LME								
[16]	0.1	1.4	23.6	14.2	0	44.2	10.7	0.4
[10]	0.1	0.1	25.5	15.8	0	47.1	8.9	1.1
[11]	1.2	1.86	24.49	14.39	2.8	38.32	13.44	3.3
RME								
[15]	0	0	4.2	2	0	64	20.6	9.2
[13]	0	0	3.5	0.9	0	64.1	22.3	8.2
[8]	0	0	3.49	0.85	0	64.4	22.3	8.23
[16]	0	0.1	4.8	0.4	0.2	61.6	20.6	9.2
[6]	0	0	4.3	1.9	0	61.5	20.6	8.3

*Explanations as in Table 1.

**1. Van Gerpen, 1996; 2. Holser and Harry-O'Kuru; 3. Monyem *et al*., 2000; 4. Imara *et al.*, in press; 5. Geller and Goodrum, 1996; 6. Kincs, 1985; 7. Neto *et al.*, 2003; 8. Ramadhas *et al.*, 2005; 9. Zullaijah *et al.*, 2005; 10. Allen *et al*., 1999; 11. Kinast, 2003; 11. Lapuerta *et al.*, 2005; 12. Demirbas, 2005; 13. Crabble *et al.*, 2001; 14. Dunn and Knothe, 2001; 15. Leung *et al.*, 2006; 16. Rakopolulos *et al.*, 2006; 17. Liang *et al.*, 2006.

T a b l e 4. Cetane number (CN) of pure fatty acid methyl esters

CN	Lauric	Mystic	Palmitic	Stearic	Palmitoleic	Oleic	Linoleic	Linolenic
61.1	100	0	0	0	0	0	0	0
69.9	0	100	0	0	0	0	0	0
74.4	0	0	100	0	0	0	0	0
76.3	0	0	0	100	0	0	0	0
51.0	0	0	0	0	100	0	0	0
57.2	0	0	0	0	0	100	0	0
36.8	0	0	0	0	0	0	100	0
21.6	0	0	0	0	0	0	0	100

of FAME. However, the range of reported values for CN of different types of biodiesel as shown in Table 1 varies considerably. The numbers in brackets are the references from where the CN values were taken. It is seen that the CN varies from 45.0 to 60.0 in SME, 48-59.7 in RME *etc*. The difference has been attributed partially to experimental error, and the probability of oxidation of the biodiesel before use which has been found to increase CN (Van Gerpen, 1996).

Table 5 shows average FAME compositions of different feedstock for biodiesel fuel as extracted from past works (Table 2). From Table 3, the distribution of FAME compounds was not the same for all the feedstocks, though a pattern was noticed from one feedstock to another. For example, linoleic acid and oleic acids are prevalent in sun-flower, cottonseed, rapeseed, peanut and soybean methyl esters. However, palmitic acid and oleic acid are predominant in palm oil and lard, while tallow is a combination of palmitic, stearic and oleic acids. These variations will certainly affect the CN of the various feedstocks.

A graphical representation of the variation of FAME composition of eight different feedstocks is shown in Fig. 1; it is seen that soybean biodiesel consists mainly of linoleic acid and oleic acid. The CN of pure linoleic acid is 36.8, while that of oleic acid is 57.2. The average CN obtained from the literature was 50.7, while the predicted value was 50.3. This is very close and represents the range of the CN of

T a b l e 5. Average % composition of fatty acids of different feedstocks

Feedstock	Lauric	Mystic	Palmitic	Stearic	Palmitoleic	Oleic	Linoleic	Linolenic
SME*	0	0.06	10.64	3.88	0.14	32.38	46.36	5.53
SUNME	0	0.01	6.48	4.25	0	18.97	69.07	0.26
PME	0.29	0.87	43.08	4.31	0.12	40.55	9.64	0.32
TME	0.08	1.3	23.88	17.88	0	45.25	2.85	0.53
CME	0.02	0.32	22.05	2.17	0.13	16.13	55.72	0.25
PEME	0	0	11.1	4.22	0	47.23	32.14	0.68
LME	0.07	1	26.03	15	0	45.43	9.87	0.5
RME	0	0.02	4.06	1.2	0.04	63.12	21.28	8.63

*Explanations as in Table 1.

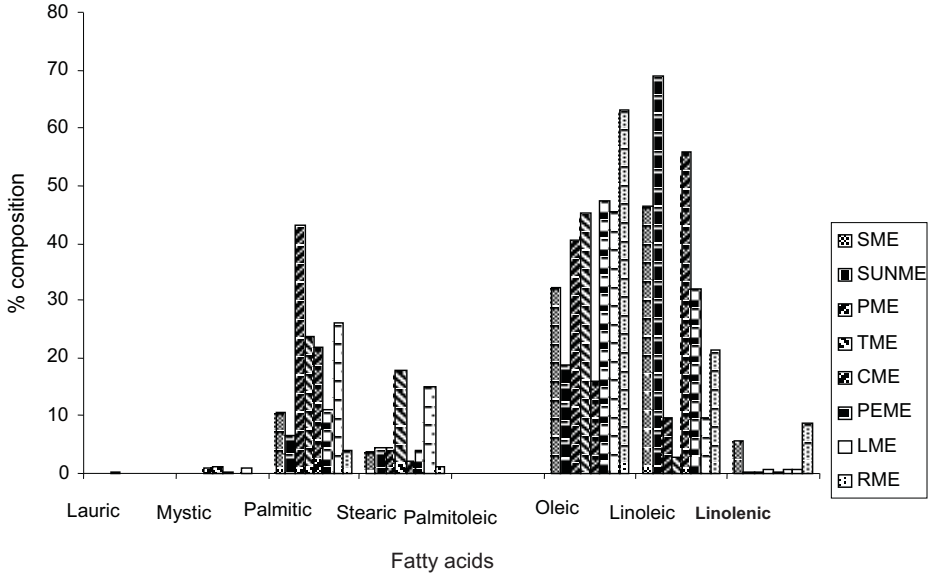

Fig. 1. Fatty acid compositions of eight feedstocks.

the dominating FAME in the biodiesel. The contributions of other acids, especially the 10% palmitic acid, will add to the increase in the CN number because of its high value of CN. Linoleic acid is predominant, close to 70% composition, oleic acid of about 20%, with the remaining acids sharing the rest in small percentages in sunflower biodiesel. The average measured value of CN was 53.9, while the predicted value was 48. It is expected that the CN should be within the range of 36.8 and 57.7, and should be closer to 36.8 than 57.7, considering that the composition of FAME is high in linoleic acid. The predicted value of 48 could be said to fall within the expected value in view of the dominant value of linoleic acid. However, it was closer to 57.7 than 36.8, which may be due to the effect of the 6% palmitic acid and 4% stearic acid which tend to increase the overall CN. The CN of these acids are 74.6 and 76.3, respectively. The measured values may then not be totally accurate. The sharp disparity here could be due to oxidation of the fuel since some reports establish an increase in CN with oxidation up to 82 value of peroxide in the biofuel (Van Gerpen, 1996).

The same trend was observed in palm oil methyl ester where palmitic acid and oleic are the major constituents of the methyl ester. This is a combination of saturated and unsaturated acids. It has been found out that CN increases with saturation and decreases with unsaturation (Van Gerpen, 1996). The CN for pure methyl oleate was 57.2, while that of pure palmitate was 74.4. The average CN from literature was 62.7, while the predicted value was 63.1. The values are within the range of the two prominent FAME constituents, and closer to each other.

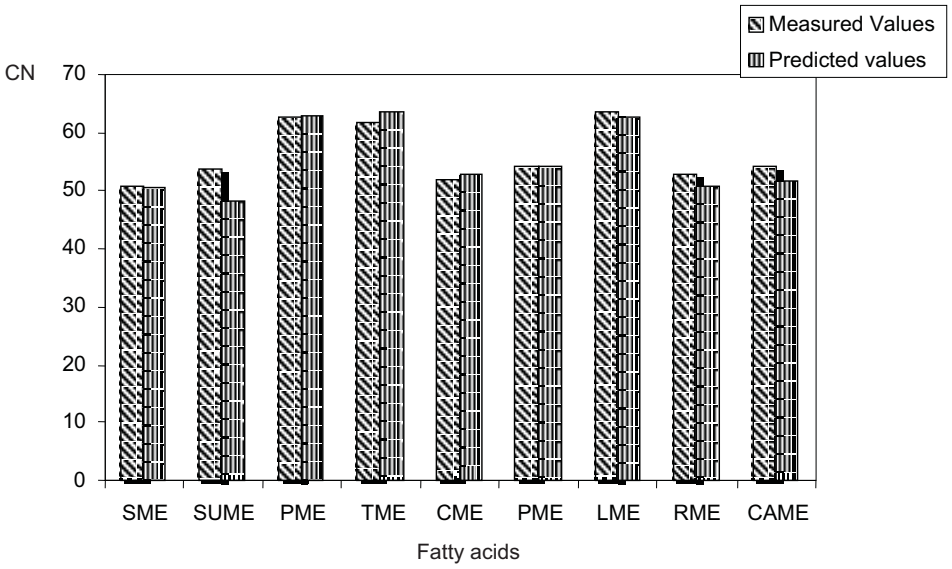

Fig. 2. Relationship between average measured and predicted values of cetane number (CN).

Linoleic acid is the predominant acid in cottonseed, with oleic and palmitic acids contributing about 16% and 22%, respectively. The average CN from literature was 52, while the predicted value was 52.8. This is close to each other and to the range of CN values of the pure FAME constituents.

The most dominant fatty acid in rapeseed is oleic acid, having about 60% of the composition, with 20% of linoleic acid. The average CN was 52.8, while the predicted value was 50.6. It is within the range of the CN of the dominant FAME. The FAME composition of peanut shows that oleic acid is predominant, representing about 47% of the composition, with linoleic acid being the second predominant with about 30% of the composition. The average CN from literature was 54, while the predicted value was 54. The value is still within the range of the two major acids in the oil. The same trend was observed in lard and tallow.

The CN follows the same trend in the composition of the FAME compounds in all the feedstocks used. It was observed that a feedstock that is high in saturated fatty esters has a high CN, while feedstocks predominant in unsaturated fatty acid have lower CN values. This agrees with a previous report by Van Gerpen (1996). This suggests a profound effect of fatty acid composition on the CN of the biodiesel.

As illustrated in Fig. 2, the average measured values of CN obtained from literature compare well with the predicted values using the generated equation. However, there are slight variations in rapeseed, sunflower, and tallow biodiesels. This may be connected with the variations in the results obtained by different researchers. The predicted values, however, fall within the range of the measured values. It has been established that a change in the oxidation level of

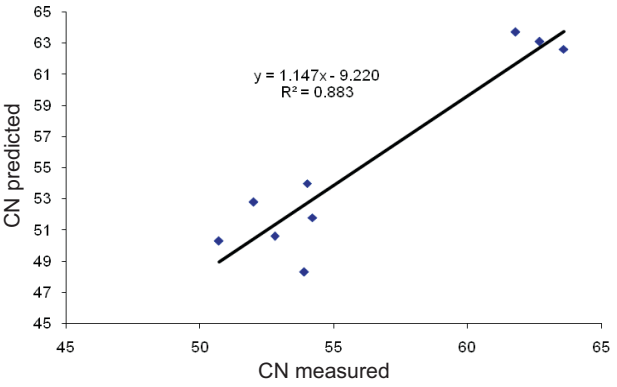

Fig. 3. Relationship between measured and predicted values of cetane number (CN).

biodiesel would have an impact on the cetane number (Vellguth, 1983). An increase in the peroxide value of the biodiesel up to 82 has been found to increase CN (Van Gerpen, 1996). This may account for the changes in CN of different stocks.

The equation developed as Eq. (2) was used to predict the CN values of different biodiesels consisting of different FAME compositions. A plot of predicted values of CN against the measured values from literature is illustrated in Fig. 3. The coefficient of determination (R^2) was 0.88, which indicates that the CN can be predicted with 88% accuracy based on the FAME composition of the biofuel. A clear pattern was observed from Fig. 3 showing two different groups of CN values, one forming the range of 48-54, and the other - 61.8 to 63.6. The CN values in the first range are

biodiesel fuels from vegetable oils, while the second range refers to biodiesel fuels from animal fats and palm oil. It was observed that FAME from vegetable oils are mostly unsaturated, while the animal fats and palm oil are saturated fatty acids. This further supports the fact that the CN is affected by the % composition of FAME, as CN values of the saturated FAME are above 60, while those of unsaturated FAME are below 60, as seen in Table 2.

CONCLUSIONS

1. It is established that the FAME composition of the methyl esters used has a predominant effect on the cetane number (CN) of the biodiesel. From the results obtained, it is evident that CN is affected by the % composition of the FAME in the fuel.

2. An equation was developed to predict the CN based on the FAME composition and it was able to predict with 88% accuracy. The developed equation can effectively predict the CN of the biodiesel based on its FAME composition.

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