Role of Internal Pressure-Free Volume in the Study of Calorific Value of Vegetable Oils as Biodiesel

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Abstract: Many studies have been made on physio-chemical properties of several vegetable oils. However, the thermodynamic properties such as internal pressure and free volume and their relation to fuel characteristics like higher heating value and Cetane number have not been made so far and are reported for the first time. The nature of the internal molecular structure and its impact on the physical and thermodynamic properties is studied and reported. Though oils have a very complex structure, their physio-chemical properties mainly depend on the average chain length of fatty acids and their level of saturation.

Keywords: vegetable oils, internal pressure, free volume, higher heating value, and Cetane number

1. Introduction

In view of protecting the global environment as well as to alleviate the crisis caused by the exhaust of fossil fuels, compound fuels derived from vegetable oils or animal fats have been studied all over the world, as alternative fuels called bio diesel . Biodiesel is generally made of (m) ethyl esters of fatty acids produced by the transesterification reaction of triglycerides with any alcohol in the presence of alkalis, acids or enzymes as a catalyst [1]. It can be used in pressure-ignited internal combustion engines or for other types of energy generation. An attempt is made in this paper to estimate a few physical properties like Iodine Value (IV), Saponification Value (SV), Higher Heating Value (HHV) and Cetane Number (CN) for 8 vegetable oils and correlate these properties to thermodynamic parameters such as internal pressure (Pi) and Free Volume (Vf) of oils calculated for the first time.

2. Theory

Vegetable oils can partially or totally replace fossil fuels with their long chain hydrocarbon structure which yields them good ignition characteristics. Also vegetable oils are ecofriendly than diesel in terms of flash point, aromatic content, sulfur content and biodegradability [2] -[7]. However the carbon deposits, excessive buildup of straight vegetable oil (SVO) in the lubricants, the lower calorific value along with high value of viscosity become some of the hindrances to choose them as better alternatives to mineral oils. Several studies are reported to improve the fuel efficiency of the vegetable oils, such as pyrolysis, dilution, transesterification etc. [8] -[11]. Chemical structure always influences the physical properties of any material. In the case of oils, chain length is reported to have positive influence on viscosity, Higher Heating Value and Cetane number; while unsaturation has negative influence on these

properties.

Flash point is positively influenced by chain length and unsaturation. Branching seems to be having little influence on many of the physical properties [12] - [13]. The studies on free volume and internal pressure in liquids throw light into the intermolecular interactions existing in them. Rate process in any liquid is determined by its molecular packing. Structure indices such as Iodine value (IV) and Saponification value (SV) are used to relate structure with the chemical properties.

2.1Viscosity and Density

Viscosity is a measure of internal fluid friction or resistance offered by the liquid to its free flow. Viscosity, density and ultrasonic velocity and allied thermodynamic parameters play a vital role in studying the intermolecular forces in liquid solutions [14] -[17]. Vegetable oils are extremely viscous compared to the diesel oil. Density is the mass per unit volume of any liquid and is one of the major parameter contributing to various physical and chemical properties [18]-[21].

2.2 Free volume

As far as liquid molecules are concerned, they are not closely packed and as such there is some free space between them. This free space in the liquid molecules, named as the free volume, is expressed in ml/mole given as per the equation [22],

$$V_{\rm f} = \left[\frac{Mu}{K\eta}\right]^{\frac{3}{2}} \tag{1}$$

Where M is the molar mass in gm., u is ultrasonic velocity in cms-1, $K=4.28 \times 10^9$, η is the viscosity in cP.

2.3 Internal pressure

Internal pressure is a fundamental property of liquid. It explains the forces of attraction and repulsion between the molecules and can be used to relate the molecular association with hydrogen bonding [23] - [28]. In the current study, the internal pressure (P_i) is calculated using the concept of the free volume as given by the equation [29]

$$P_{i} = \frac{bRT}{(V^{2}V_{f})^{\frac{1}{3}}}$$
(2)

Where b=1.78, R is the universal gas constant, V is the molar volume and V_f the free volume.

2.4 Calorific value

Calorific value or heat of combustion gives the energy content in the fuel as well as the fuel efficiency. It is the amount of energy released per unit quantity by the fuel during the combustion process [30] -[33]. Higher Heating Value (HHV) of the oil, which is the amount of heat evolved when one mole of compound is burned to CO_2 and $H_2 O$; can be obtained either from the physical properties of the oils, such as viscosity (VS), density, flash point or from their fatty acid composition. In this study HHV is obtained from viscosity, using the relation given below [34].

$$HHV = 0.0317 VS + 38.053$$
(3)

2.5 Iodine value (IV)

Fatty acid composition of vegetable oils is a mixture of Saturated Fatty Acids (SFA) and Unsaturated Fatty Acids (UFA). Iodine Value (IV) indicates the average amount of unsaturation of the oils [35] -[36]. IV of a mixture of fatty compounds can be calculated from the following equation [37],

$$IV = \sum 100 \times \frac{A_{f} \times 253.81 \times db}{MW_{f}}$$
(4)

Where A_f is the amount in percentage of a fatty compound in a mixture, db is the number of double bonds and MW_{\sharp} is the molecular weight of the fatty compound. As it is the measure of the degree of unsaturation of the fuel, it will provide the information about deposit problems and storage stability problems with the fuels.

2.6 Saponification value (SV)

Saponification value gives the amount of fatty acids either free or bound to the glycerol of a vegetable oil. The estimation of saponification value in vegetable oils is done using the fatty acid profile as given below [38].

$$SV = \sum \frac{A_f \times 56106}{MW_f}$$
(5)

Where A_f the amount in percentage of a fatty compound in a mixture and MW_f is the molecular weight of the fatty compound.

2.7 Cetane number (CN)

CN is the measure of ignition quality; the ignition delay time of the fuel. Cetane number is affected by the number of carbon atoms of the original fatty acids, the number of double bonds, viscosity etc. The longer the fatty acid carbon chains and the more saturated the molecules, the higher is the Cetane number. The determination of Cetane number involves a series of laboratory experiments, involving considerable energy and time. Hence predictive equations have been developed by various researchers to account for the viscosity and Cetane number of various oils, using the triglyceride constituents in the oils [39]-[40].

The calculation of the Cetane number using regression analysis making use of fatty acid profile is used in this work [41] as expressed below,

 $\begin{array}{c} \text{CN}=61.03+0x_1+0.1025x_2+0.133x_3+0.152x_4-0.0\\ 01x_5-.037x_6-0.243x_7-0.395x_8 \quad (6)\\ \text{Where } x_1\dots x_8 \text{ are percentage of fatty acid composition.} \end{array}$

The Cetane number (CN) is a dimensionless descriptor for the ignition delay (ID) time of a diesel fuel upon injection into the combustion chamber.

2.8 Calculation of Oxidative Stability (COX) of Oils

The COX value of oils was calculated by the percentage of C18 unsaturated fatty acids, using the formula proposed by

$$\text{OX value} = \frac{1 \times x_1 + 10.3 \times x_2 + 21.6 \times x_3}{100}$$
 (7)

where x_1 , x_2 , and x_3 is the percentage of $C_{18:1}$, of $C_{18:2}$, and of $C_{18:3}$ in total fatty acids of an oil sample, respectively [42].

The COX value can be viewed as the measure of tendency of oils to undergo autoxidation. It was reported that vegetable oils with high contents of $C_{18:2}$ and $C_{18:3}$ fatty acids were susceptible to oxidation, and presence of SFA and MUFA could improve oxidative stability. Oxidation of unsaturated esters in biodiesel occurs by contact with air and other pro-oxidizing conditions during long term storage. Thus, oxidative stability is an important issue that biodiesel research must address since oxidation product may impair fuel quality and subsequently, the engine performance [43]-[44].

3. Material and Methods

Commercial vegetable oil samples from reliable and standard manufacturers were procured. The oils were filtered and equilibrated at 25° C for 24 hours before measurement. A minimum of four samples were used for all the measurements. Density was calculated by measuring mass of 10 samples of each oil using a R.D bottle and a METLAR electronic balance, correct to 0.001mg. The mean value after comparison with the literature is reported in gm cm⁻³.

Viscosity was calculated by measuring the time of flow using an Ubbelohde viscometer. A stop watch with an accuracy of 0.01s was used. A minimum of ten trials were made and the average value is calculated and reported in cP. An ultrasonic interferometer (Mittal enterprises New Delhi, India) was used to find the ultrasonic velocity at 2MHz. A minimum of four trials have been made and the mean value of velocity is reported inm.s⁻¹.

An overall observation on the measurement is that the measured and the computed parameters showed minimal changes (less than 2%) with storage time up to one month. All graphs were plotted using ORIGIN 8.1 software and the statistical parameters were computed using the same package.

4. Result and Discussions

For the 8 oils studied, the measured values of density, viscosity and ultrasonic velocity are given in Table 1.

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Name of	Density	Viscosity	Velocity	Molar mass	Molar				
Oil					Volume				
	gm.cm ⁻³	cР	$m.s^{-l}$	gm.	ml/mole				
CRN	0.920	65	1470	868	943				
OLV	0.916	92	1464	830	906				
CTTN	0.919	51	1418	870	947				
PEA	0.913	74	1418	885	969				
PLM	0.919	85	1459	840	914				
CST	0.899	30	1475	927	1031				
COC	0.964	40	1420	650	674				
JAT	0.900	53	1433	800	879				

 Table 1: Measured physico-chemical parameters of 8

 vegetable oils

*CRN-CORN OIL, OLV-OLIVE OIL, CTTN-COTTON SEED OIL, PEA-PEANUT OIL, PLM-PALM OIL, CST-CASTOR OIL, COC-COCONUT OIL, JAT-JATROPHA

Based on the values of density, viscosity and ultrasonic velocity, free volume and internal pressure is calculated. Using the equations given earlier, calculations were also made for HHV, IV, SV, CN which are given in Table 2.

Table-2: Free volume-internal pressure values along with other structural indices of 8 Vegetable oils

Name	Vf (ml/	Pi	HHV	IV	SV	CN	UFA/	COX
of oil	mole)	(atm)	(MJ/Kg)		1		SFA	value
CRN	0.010	2175	40.7	133.8	197.8	46.8	6.0	6.4
OLV	0.005	2734	41.6	83.3	198.2	58.3	5.6	1.9
CTTN	0.013	1962	40.3	118.3	199.7	49.9	2.5	5.9
PEA	0.008	2307	41.0	62.9	138.5	57.9	4.4	3.9
PLM	0.006	2601	41.3	55.2	205.0	63.4	1.1	1.5
CST	0.035	1345	39.6	90.5	198.9	55.9	35.5	1.5
COC	0.013	2519	39.9	10.0	222.5	64.2	0.1	0.3
JAT	0.011	2181	40.3	108.7	198.9	52.3	3.6	4.1

It may be observed from the table that castor oil having the lowest viscosity possesses the highest free volume, contributing to the lowest internal pressure. Olive oil has the highest viscosity indicating the tight package of molecule which in turn gives it high internal pressure and high calorific value (Table 2). Also it is been observed that out of the 8 vegetable oils under study, castor oil is having the highest free volume, leading to the lowest internal pressure and calorific value.

The measured parameters of the oils in this study showgood agreement with the literature values [45], 46]. Viscosity of the vegetable oils is found to have a direct correlation with .67.

the carbon chain length and the amount of fatty acids. The viscosity tends to decrease when there is an increased presence of double bonds and grows with an increase in the length of the hydrocarbon chain and according to the level of polymerization in the oil. Higher the value of CN, more fuel-efficient the fuel becomes. Lower iodine value implies lower content of unsaturated fatty acids and low saponification value can be because of higher saturated fatty acid compounds. As the value of CN increases, fuelefficiency also increases. It may be seen that castor oil has a predominantly high content of fatty acids (94%) This is reflected in its lowest COX value leading to slow oxidation and high resistant to rancidity. Because of this, castor oil can be used as feedstock in the production of biodiesel. Also other properties of castor oils are also distinctly different. The relation between internal pressure and various physicchemical properties of vegetable oils such as HHV, IV, SV and CN is given in the table (3).

Table 3: relation between Pi with HHV, IV, SV and CN

Graph	Correlation equation	R^2				
between		value				
Pi with	$y = b_0 + b_1 x + b_2 x^2$	0.825				
IV	$b_1 = -0.00757, b_2 = -9.69E-6$					
Pi with	$(-2(\frac{x-x_c}{x_c}))^2$	0.82				
SV	$y = y_0 + (A(w^* \sqrt{\frac{2}{2}} e^{(w^* y^*)}))$					
	$v_0 = 204.83$, $x_c = 2291.69$, $w = 103.85$, $A = -$					
\backslash	9017.26					
Pi with	$v = v_0 + (A * e^{(-0.5(\frac{x - x_c}{w}))^2})$	0.60				
CN	$y_0 = 60, x_c = 2072, w = 114.99, A = -16.7$					
Pi with	$y = b_0 + b_1 x + b_2 x^2$	0.98				
HHV	$b_1 = 9.05E-5$, $b_2 = 2.69E-7$					

The relation between the internal pressure and the free volume in various vegetable oils is shown in the fig. (1). An inverse relation as expected is obtained between internal pressure and free volume with R^2 value as 0.92. A distinct observation is the deviation shown by the coconut oil.



Figure 1: variation of internal pressure with free volume

The variation of the internal pressure with the stearic acid content is given in the figure (2). In the present study the internal pressure shows an exponential increase with the stearic acid ($C_{(18:0)}$) content with R2 value as 0



Figure 2: Variation of Pi with stearic acid content.

The variation of the internal pressure with the UFA/SFA ratio is shown in figure (3). The internal pressure in the vegetable oils under study shows an exponential decrease the level of saturation with R2 value as 0.97.





The variation of the calorific value with respect to the PUFA content in the veg oils is shown below in figure (4).



Figure 4: variation of HHV with PUFA content.

The plot showing the variation of HHV with PUFA content is showing a second order polynomial fit with R2 value of 0.99

5. Conclusions

The role of internal pressure and free volume in relation to the fuel characteristics has been studied for 8 vegetable oils and reported for the first time. The importance of molecular interactions in calculating these properties is also highlighted. It is found that the internal pressure in the study shows a relation with the stearic acid content in the vegetable oils. Further studies have to be conducted in this regard for details. Also the calorific value of the vegetable oils exhibits a second order polynomial relation with the PUFA content of the oils. Detailed investigation has to be done in this regard also. As the thermodynamic properties and molecular structure contribute to the improvement of technology used for energy transformation, this may lead to new technological solutions for planning suitable plant for biodiesel production.

6. Acknowledgement

The authors are thankful to Prof. J.F. Rajasekaran, retired professor of Physics, Madras Christian College Tambaram, for helping with experimental measurements and computation. The authors also acknowledge the help and encouragement given by Dr. Thomas P John, Chairman of T John group of institutions and Dr. H.N. Thippeswamy, Principal of T John Institute of Technology. Also we greatly admire the guidelines given by Dr. Hina G Oza during the investigation.

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Volume 6 Issue 2, February 2017 www.ijsr.net

International Journal of Science and Research (IJSR) ISSN (Online): 2319-7064 Index Copernicus Value (2015): 78.96 | Impact Factor (2015): 6.391

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Volume 6 Issue 2, February 2017

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