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Complementary Blended Sustainable Hybrid Biodiesel for Fuel Property Enhancement: A Review

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***ABSTRACT**: Biodiesel in spite of its suitability as diesel fuel alternative suffers from poor oxidative stability and poor cold flow properties and higher Nox emissions as compared to diesel which poses difficulties in storage and utilization as fuel. As the properties of biodiesel largely dependent on fatty acid profile of the feedstock which contains different fatty acids in different proportions, one feedstock cannot satisfy all the requirements of ideal biodiesel composition. Different techniques applied for improving cold flow properties and oxidative stability of triglyceride fuel but complementary blending of biodiesels is low cost, simple and effective method. Biodiesels were blended in different weight ratios and their properties like oxidation stability, kinematic viscosity and cold flow properties were measured. Most of the blended biodiesel satisfied biodiesel standards and for some blends properties improved but blend failed in satisfying one of the standard parameters of biodiesel. As per the local availability combination of non edible and waste oils like waste cooking/frying oil, acid oils, used transformer oil and animal fats can be utilised for production and blending of biodiesel to reduce the cost of biodiesel and prevent food vs fuel scenario.*

Key words: FA- fatty acid; ME- methyl ester; BD- biodiesel; OS-oxidation stability; IP- induction period; CFPP- cold filter plugging point; CP- cloud point; PP- pour point; KV-kinematic viscosity; IV- iodine value

1. INTRODUCTION

Biodiesel is a mixture of mono alkyl esters with long-chain fatty acids that is derived from vegetable oils or animal fats[1]. Biodiesel in spite of its suitability as diesel fuel alternative suffers from poor oxidative stability and poor cold flow properties and higher Nox emissions as compared to diesel which poses difficulties in storage and utilization as fuel[2]. In order to find the ideal biodiesel composition for optimum engine performance and emission, the chemical composition of biodiesel plays an important role [2]. Suitable proportions of saturated and unsaturated fatty acids in feedstock are required to attain improved cold flow performance and higher oxidation stability in biodiesel[3].

In search of the ideal biodiesel composition, Greater amount of monounsaturated fatty acids lower amount of polyunsaturated acids and controlled presence of saturated acids are required for ideal biodiesel composition. are the best-fitting acids In terms of oxidative stability and cold weather behaviour, presence of C18:1 and C16:1 are strongly recommended [4]. As the properties of biodiesel largely dependent on fatty acid profile of the feedstock which contains different fatty acids in different proportions, one feedstock cannot satisfy all the requirements of ideal biodiesel composition.

Different techniques applied for improving oxidative stability and cold flow performance of triglyceride fuel are mixing of diesel and oil in different proportion[5], winterization and use of cold flow additives[6], modifying the fatty acid structure of biodiesel by genetic modification[7], isomerisation over solid acids [8] and complementary blending of biodiesels having suitable ester profile[9, 15-19]

This first of its kind review paper is aimed to focus on the relatively simple but effective method of complementary blending of biodiesels produced from different feedstocks to improve the biodiesel properties and its engine performance and emission results to arrive at the optimum blend of biodiesels.

2. BIODIESEL FEEDSTOCKS AND COMPOSITION

Biodiesel fatty acid composition corresponds to that of its oil feedstock and proportion of different fatty acids present in biodiesel varies significantly as per the feedstocks. [7]. Table 1 shows chemical structure of common fatty acids found in feedstocks which can be categorized as saturated or unsaturated according to presence of number of double bonds In carbon chain.

Table 1. chemical structure of common fatty acid[14]

fatty acid name	Chemical name	Fatty acid chain structure (xx:y)	Chemical formula	Saturated/Unsaturated
Lauric	Dodecanoic	12:0	$C_{12}H_{24}O_2$	Saturated
Myristic	Tetradecanoic	14:0	$C_{14}H_{28}O_2$	Saturated
Palmitic	Hexadecanoic	16:0	$C_{16}H_{32}O_2$	Saturated
Stearic	Octadecanoic	18:0	$C_{18}H_{36}O_2$	Saturated
Arachidic	Eicosanoic	20:0	$C_{20}H_{40}O_2$	Saturated
Behenic	Docosanoic	22:0	$C_{22}H_{44}O_2$	Saturated
Lignoceric	Tetracosanoic	24:0	$C_{24}H_{48}O_2$	Saturated
Oleic	cis-9-Octadecenoic	18:1	$C_{18}H_{34}O_2$	Mono-unsaturated
Linoleic	cis-9,cis-12-Octadecadienoic	18:2	$C_{18}H_{32}O_2$	Di-unsaturated
Linolenic	cis-9,cis-12,cis-15-Octadecatrienic	18.3	$C_{18}H_{30}O_2$	Polyunsaturated
Erucle	cis-13-Docosenoic	22:1	$C_{32}H_{42}O_2$	Mono-unsaturated

(xx:y) = (the number of carbons: the number of double bonds) in the fatty acid chain

For satisfactory working of liquid transportation fuels, phase change from liquid to solid at lower temperature is objectionable. The melting point (mp) of fatty acid alkyl esters largely dependent on number of double bonds, hydrocarbon chain length, type of ester, and location [15]. Melting point(mp) of various fatty acids present in biodiesel shows trend of increase in mp with increase in hydrocarbon tail length[15].

The CP is the temperature at which a liquid fatty material becomes cloudy due to formation of crystals and solidification of saturates. Solids and crystals rapidly grow and agglomerate, clogging fuel lines and filters and causing major operability problems. Pour point of liquid is the lowest temperature at which it will still flow with formation of solids . Biodiesel being a mixture of saturated and unsaturated fatty esters, it crystallize at higher temperature because of presence of saturated fatty acids which exhibit higher mp than unsaturated fatty acids. Selection of potential oil for biodiesel production in terms of availability, Cold flow properties and oxidative stability concerns optimum combination of saturated and unsaturated fatty acids without any cold flow improvers and additives.

3. FUEL PROPERTY ENHANCEMENT USING COMPLEMENTARY BLENDS OF BIODIESELS

Biodiesel Fuel properties are dependent on the Fatty Acid composition of the source oil or fat from which biodiesel is produced. Therefore, by using complementary blending of biodiesel can reduce high feedstock cost and fuel properties can be enhanced for better performance as fuel[3,11,13,16]. As a result, to examine the effect of complementary blending of biodiesel on fuel properties, exploration of blends is important.

Different authors studied about complementary blended biodiesels sourced from various types of feedstocks to compensate the poor cold flow property , poor oxidation stability and higher viscosity of specific biodiesel by selecting feedstocks having one of the superior properties.

Bryan R. Moser [11] argued that camelina, cottonseed, palm and soybean oil-derived biodiesels exhibited poor oxidative stabilities but satisfactory kinematic viscosities. Field pennycress and meadowfoam seed oil methyl esters yielded excellent cold flow properties but high kinematic viscosities. Thus, fuels were blended with camelina, cottonseed, palm and soybean oil-derived biodiesels were blended with field pennycress and meadowfoam derived biodiesel which have better oxidative stabilities and excellent cold flow properties but higher kinematic viscosities to ensure the improvement in selected properties. Table 4 shows important properties of biodiesels and its fatty acid profile and Table 5 shows properties of biodiesel blended in different ratios.

Table 2. Important properties of biodiesels and its fatty acid profile

Properties/Bio-diesel	CSME	CTME	FPME	MFME	PME	SME	Reference
CP (°C)	4.1	5.6	-6.5	-6.6	15.3	0.4	[11]
CFPP (°C)	0.0	6.3	-18.7	-10.0	12.3	-3.0	
PP (°C)	0.0	5.0	-24.3	-10.0	14.0	-1.0	
IP (h)	2.7	3.7	2.4	67.3	13.9	3.8	
KV (mm^2/s)	4.37	4.19	5.49	6.22	4.53	4.10	
Fatty acid composition							
Saturated FA(%)	11.6	28.1	5.8	1.9	51.1	15.4	
Mono un-Saturated FA (%)	33.3	17.9	61.7	78.1	40.6	23.6	
Polyun-Saturated FA (%)	52.6	53.3	30.0	18.0	10.5	60.3	

Table 3. Measured Properties of biodiesel blended in different ratios

Sr.No	Blending ratio(vol %)	CP (°C)	CFPP (°C)	PP (°C)	IP(h)	KV (mm^2/s)	Reference
1	CSME:FPME (25:75)	-2.8	-13.3	-21.0	2.7	5.15	[11]
2	CTME:FPME (25:75)	-8.5	-9.0	-12.3	3.0	5.13	
3	PME:FPME (25:75)	0.5	-2.7	-6.3	3.4	5.20	
4	SME:FPME (25:75)	-5.9	-17.0	-17.3	3.5	5.10	
5	CSME:MFME (25:75)	-5.4	-7.7	-9.0	12.2	5.72	
6	CTME:MFME (25:75)	-5.0	-8.0	-13.0	12.7	5.63	
7	PME:MFME (25:75)	-1.1	-4.7	-6.3	39.2	5.70	
8	SME:MFME (25:75)	-7.2	-10.0	-8.7	12.5	5.57	

FPME had CP of -6.5 ºC, PP of -24.3 ºC and CFPP of -18.7 ºC and and MFME had CP of -6.6 ºC, PP of -10.0 ºC and CFPP of -10.0 ºC which were lower than CSME, CTME, PME, and SME. The Kinematic Viscositys of FPME was 5.49 mm^2/s that of MFME was 6.22 mm^2/s which were not satisfactory with the limits prescribed in the biodiesel standards. Complementary blending of MFME with other selected methyl esters can improve cold flow properties of CSME, CTME, PME, and SME while simultaneously lowering the KVs of FPME and MFME. Author derived statistical regression generated equations for predicting fuel properties from known blend ratios. Such equations could be highly accurate for prediction of KV was accurate but prediction of cold flow properties was reasonable. Oxidative stability prediction was not accurate at all.

Ji-Yeon Park et al.[17] tried to improve the oxidation stability and the cold flow properties of a biodiesel by blending three different kinds of biodiesels palm, rapeseed, and soybean in different weight ratios. He also checked effect of fatty acid compositions on oxidation stability and the cold filter plugging point (CFPP).

Table 4. Properties of raw biodiesels and its blends in different ratios and its fatty acid profile as per saturation

Sr. No	Blending ratio(wt%) (palm:rapseed:soybean BD)	Oxidation stability(h)	CFPP (ºC)	Palmitic Acid (wt%)	Oleic acid (wt%)	Linoleic acid (wt%)	Linolenic acid (wt%)	Reference
				Saturated	Unsaturated			[17]
1	0:0:100	3.87	-3	11.0	23.1	53.3	6.8	
2	0:100:0	6.94	-20	4.4	62.4	19.7	9.5	
3	100:0:0	11.00	10	40.1	43.0	11.0	0.2	
4	20:60:20	6.56	-6.0	12.9	50.7	24.7	7.1	
5	40:40:20	7.65	-3.0	20.0	46.8	23.0	5.2	
6	60:20:20	7.97	3.0	27.1	42.9	21.2	3.4	

For 20:60:20 ratio of palm, rapeseed, and soybean biodiesels , the oxidation stability of the blended biodiesel was 6.56 h and its CFPP was -6.0 ºC. which shows improved OS of blend due to presence of palm and rapeseed biodiesels having higher oxidation stability. By blending palm biodiesel with rapeseed and soybean and rapeseed biodiesels, CFPP of palm biodiesel was also improved. To show reliance of oxidation stability and cold flow properties on saturation value of FAME, different correlations were obtained. If the compositions of the blended biodiesels are determined, it is possible to predict their oxidation stability and CFPP.

Atabani, A. E. et. Al. [18] used different oils for biodiesel preparation and blending which include Jatropha curcas, Calophyllum inophyllum, Sterculia foetida, Moringa oleifera, Croton megalocarpus, Patchouli, Elaeis guineensis (palm), Cocos nucifera (coconut), Brassica napus (canola) and Glycine Max (soybean). Blending of SFME and CoME improved the viscosity of SFME from 6.3717 mm^2/s to 5.3349 mm^2/s for 3:1 blend, 4.4912 mm^2/s for 1:1 blend and 3.879 mm^2/s for 1:3 blend. The polynomial curve fitting method was used to measure other biodiesel bend properties.

Amit Sarin et. Al. [13] examined the blending of pongamia and jatropha biodiesel with palm biodiesel for improved cold flow properties of palm biodiesel. Dependence of CP and PP on esters of fatty acid composition was also examined. Author developed correlations between cold flow properties and palmitic acid methyl ester (PAME).

Table 5. Properties of raw biodiesels and its blends in different ratios and its fatty acid profile as per saturation

Sr.No	Blending ratio(wt%) (Palm:Jatropha:Pongamia BD)	CP (°C)	PP (°C)	Palmitic Acid (wt%)	Oleic acid (wt%)	Linoleic acid (wt%)	Linolenic acid (wt%)	Reference
				Saturated	Unsaturated			
1	0:0:100	-1	6	9.8	72.2	11.8	--	[13]
2	0:100:0	4.00	-3	14.2	43.1	34.4	--	
3	100:0:0	16.00	12	40.3	43.4	12.2	--	
4	20:80:0	-2	4	19.5	43.2	29.9	--	
5	80:20:0	2.0	-6.0	35.1	43.3	16.6		
6	0:20:80	0.0	-6.0	10.7	66.4	16.3	--	
7	20:20:60	2.0	-6.0	16.8	60.6	16.4		

PBD and JBD were blended in different weight ratio as shown in table 7. Blend 20:20:60 (Palm:Jatropha:Pongamia BD) had CP and PP were 2 °C and -6 °C respectively. FAME compositions of twenty one biodiesel blends of Palm, Jatropha and Pongamia biodiesels had been determined by author and some of them is given in table . Author also plotted graphs of Palmitic acid methyl ester v/s Cloud and pour point and from graphs they developed correlation between CP , PP and PAME contents (wt%).

Moser et Al. [19] complementary blended cold-pressed meadowfoam seed oil methyl esters (MFME) with soybean and waste cooking oil methyl esters (SME and WCME) to improve cold flow properties, oxidation stability, iodine value and kinematic viscosity. MFME had induction period (IP) of 66.2 h whereas SME and WCME had lower values of 5.9 and 4.5 h, respectively. The kinematic viscosity (KV) of MFME was 6.15 mm^2/s which does not satisfy biodiesel standards due to its content of unique longer-chain fatty acids. The iodine value (IV) of SME was 132 which is above the maximum limit specified in EN 14214. Lastly, the cold flow properties of MFME were superior to those of SME and WCME. From table 8 blends containing 30 vol % MFME in WCME provided IP of 8.6 h, KV of 4.92 mm^2/s and IV of 107 which satisfy both the biodiesel standards. Blend containing 50% MFME with SME and WCME satisfy both biodiesel standards. In addition, highly linear correlations between percentage of MFME and fuel properties were established. Blends were stored for 3 months and its properties were measured after storage to check the influence of storage.

Table 6. Measured Properties of raw biodiesels and its blends in different ratios

Sr.No	Blending ratio(vol %)	CP (°C)	IV(g I_2/100 g)	PP (°C)	IP(h)	KV (mm^2/s)	Reference
1	MFME:SME (100:0)	-5.8	87	-10.7	66.2	6.15	[19]
2	MFME:SME (0:100)	0.1	132	-2.0	5.9	4.05	
3	MFME:SME (20:80)	-1.5	123	-4.0	6.3	4.42	
4	MFME:SME (30:70)	-2.4	119	-5.3	7.4	4.60	
5	MFME:SME (50:50)	-4.5	110	-11.7	9.3	4.99	
6	MFME:WCME (0:100)	-0.7	116	-2.7	4.5	4.52	
7	MFME:WCME (20:80)	-1.6	110	-4.7	6.7	4.71	
8	MFME:WCME (30:70)	-2.6	107	-7.3	8.6	4.92	
9	MFME:WCME (50:50)	-4.6	102	-12.0	12.4	5.31	

Bryan R. Moser [20] complementary blended sunflower, palm, soybean and canola oil methyl esters (SFME, PME, SME, and CME, respectively) seeking improvement in KV, iodine value, cold flow properties and oxidation stability. The mixtures were prepared and properties were measured as per table 9.

Table 7. Measured Properties of raw biodiesels, its blends in different ratios and presence % of saturated fatty acids

Sr.No	Blending ratio(vol %)	OSI (h)	CP (ºC)	PP (ºC)	CFPP (ºC)	IV	SFAME	Reference
1	CME (100)	6.4	0	-9	-7	110	7.7	[20]
2	PME (100)	10.3	17	15	12	54	48.2	
3	SME (100)	5.0	1	0	-4	134	14.6	
4	SFME (100)	6.2	5	-2	2	85	9.8	
5	SME:CME (1:3)	5.9	0	-5	-8	116	9.6	
6	SME:PME (1:3)	7.7	12	13	9	73	40.9	
7	SME:SFME (1:3)	6.4	3	-1	0	97	11.1	
8	CME:PME (1:3)	9.6	11	12	8	68	38.0	
9	CME:SFME (1:3)	6.8	4	-4	0	91	9.1	
10	PME:SFME (3:1)	9.2	11	12	9	62	38.6	
11	SME:CME:PME (1:1:1)	5.4	5	5	1	99	24.0	
12	SME:CME:SFME (1:1:1)	5.0	2	-3	-6	110	10.9	
13	SME:SFME:PME (1:1:1)	6.7	5	5	1	91	24.3	
14	CME:SFME:PME (1:1:1)	7.8	4	3	-1	83	22.1	
15	SME:CME:PME:SFME (1:1:1:1)	5.7	3	2	-2	96	20.2	

Oxidation stability was improved and satisfied the biodiesel standards by blending SME with SFME. The improvement in OSI of SME through blending was attributed to the reduction in polyunsaturated FAME content of the blends in comparison to neat SME.

Zuleta et al. [21] aimed to develop different dual biodiesel blends as per table 10 with palm, castor, jatropha and sacha-inchi to improve cold flow property and induction time. Besides, dependence of these properties on saturation were also investigated.

Table 8. Measured Properties of raw biodiesels and its blends in different ratios and its fatty acid profile as per saturation

Sr.No	Blending ratio(wt%) (palm:castor:jatropha:sacha-inchi BD)	Oxidation stability(h)	CFPP (ºC)		MUME	PUME	Reference
				Saturated	Unsaturated		
1	100:0:0:0	8.85	14	48.45	41.31	10.24	[21]
2	0:100:0:0	31.72	-7	1.96	92.74	5.30	
3	0:0:100:0	3.28	1	28.24	35.65	36.48	
4	0:0:0:100	0.24	-10	6.35	8.09	85.56	
5	0:75:25:0	7.56	-12	8.53	78.47	13.10	
6	25:75:0:0	17.91	1	13.58	79.88	6.54	
7	50:50:0:0	10.78	4	25.21	67.03	7.77	
8	75:25:0:0	10.1	8	36.83	54.17	9.00	

From table 10 blend containing C75/J25 gave induction time of 7.56 h and a CFPP of −12 °C. This blend contains 8.53% saturated fatty acids , 13.10 % polyunsaturated fatty acids and 78.47% monounsaturated fatty acids. However, this blend fails to satisfy viscosity standards due to higher proportion of castor biodiesel. Blend containing P25/C75 gave very high induction time of 17.91 h and a CFPP of 1 °C as both biodiesels has good oxidation stability. To satisfy biodiesel standards, proportion of castor biodiesel should be low in the blend.

Crops et al. [22] prepeared binary mixtures castor oil biodiesel (COB), palm oil biodiesel (POB) and diesel fuel to improve viscosity, cloud point and flash point of the blends. Table 11 shows that POB:COB (80:20) blend has viscosity 5.61 mm^2/s which is below the upper limit of 6 mm^2/s but for more than 20% castor biodiesel yielded viscosity higher than 6 mm^2/s and failed to satisfy biodiesel standards. Author used thermodynamic equations and empirical models for prediction of diesel-biodiesel blend properties.

Table 9. Measured Properties of raw biodiesels and its blends in different ratios

Sr.No	Blending ratio(vol %)	CP (K)	FP (K)	KV (mm^2/s)	Reference
1	POB:COB (100:0)	289.15	424.86	4.65	[22]
2	POB:COB (0:100)	261.15	558.86	14.77	
3	POB:COB (80:20)	285.15	492.86	5.61	
4	POB:COB (60:40)	283.15	516.86	6.64	
5	POB:COB (40:60)	279.15	544.86	8.48	
6	POB:COB (20:80)	274.15	554.86	11.24	
7	Diesel:POB:COB (80:16:4)	270.15	Not measured	Not measured	
8	Diesel:POB:COB (90:2:8)	271.35	Not measured	Not measured	

By blending 20 vol.% COB with POB, flash point increased from 424.86 to 492.86 K as compared to POB. palm oil biodiesel–castor oil biodiesel (POB–COB) blends cannot be used to improve both cloud point and viscosity.

Sarin et al. [3] suggested blend of jatropha and palm biodiesel as an optimum mix for Asia. Jatropha biodiesel has good cold flow properties and palm biodiesel has good oxidation stability. Blending of these biodiesel had capability to improve both these properties as compared to individual biodiesel The blending of 20% Palm biodiesel in Jatropha biodiesel increases cloud point by 2 ºC, pour point by 3 ºC and CFPP by 1 ºC. Author blended palm biodiesel in 20,40,60 and 80 vol % with jatropha biodiesel and measured oxidation stability of the blends. He found that blends containing 60 % or more palm biodiesel in the blends gave oxidation stability more than 6 h. It is possible to attain requisite oxidation stability of biodiesel By blending 40% Palm biodiesel in Jatropha biodiesel, oxidation stability was improved with 25 ppm of antioxidant dosage. This optimum combination reduced the cost of biodiesel substantially, by use of cheaper raw material palm and needed lower quantity of antioxidant.

Chen et al. [23] used soap-nut oil biodiesel with high content of mono unsaturated fatty acid and exhibits superior oxidation stability and blended it with jatropha biodiesel to investigate CFPP and oxidation stability of the blends.

Table 10. Measured Properties of raw biodiesels and its blends in different ratios

Sr.No	Blending ratio(vol %)	CFPP (ºC)	IP (h)	Reference
1	JME:SNME (100:0)	-2	3	[23]
2	JME:SNME (0:100)	6	15	
3	JME:SNME (80:20)	-1	6	
4	JME:SNME (60:40)	-1	6	
5	JME:SNME (40:60)	3	7	
6	JME:SNME (20:80)	5	8	

For 35 wt.% or higher SNME in SNME–JME blend, the oxidation stability was observed to be greater than 6 h. With increasing proportion of SNME in SNME–JME blend, IP improves but cold flow performance deteriorates so blending with upto 40 % of SNME is recommended.

Conclusions:

The objective of this study was to present both the fuel property enhancement using complementary blended hybrid biodiesel and its performance and emission investigation. Targeted fuel properties for improvements were kinematic viscosity, oxidation stability and cold flow properties using blending of biodiesels sourced from oils having suitable fatty acid profile. Study suggested that most of the complementary blended biodiesels were at par with biodiesel standards and can be used with diesel in CI engines. Most of the complementary blended biodiesels had combination of edible and non edible oil based biodiesel. Countries which import edible oils for its domestic requirements, cannot afford to produce biodiesel from edible oils. Even though it is affordable, it also converts food into fuel so it should be taken into consideration while selecting feedstocks for biodiesel. As per the local availability combination of non edible and waste oils like waste cooking/frying oil, acid oils, used transformer oil and animal fats can be utilised for production and blending of biodiesel to reduce the cost of biodiesel and prevent food v/s fuel scenario. To increase the portion of bio fuel in partially replacing diesel, addition of bio-ethanol in complementary blended hybrid biodiesel can also be tested. If blended biodiesels sourced from multi feedstocks satisfy the biodiesel standards, it has potential to cope up the requirement of the biodiesel to partially replace the diesel. Most of the complementary blended hybrid biodiesel were tested for improvements in important properties but their performance and emission data in CI engine was not done. Very few data pertaining to only performance and emission of blended biodiesel in CI engine is available so greater insight into it is required to be researched.

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