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# *Oleo* Chemistry and Combustion Characteristics of *Plectranthus mollis,* syn. *Plectranthus incanus,* Seed Oil

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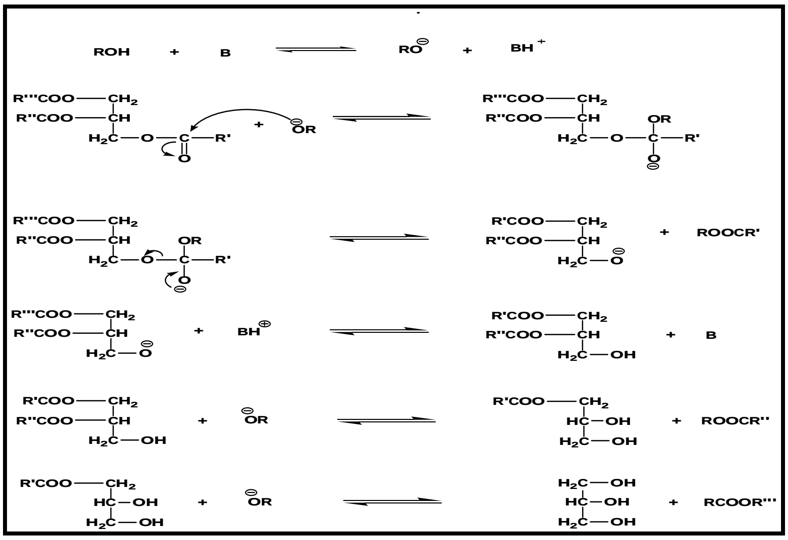
## ABSTRACT

*In this work new feedstock for biodiesel production has been screened. Non- edible seed oil species like Plectranthus mollis (PM) plant species yield 40% seed oil. The molecular weight (MW) of oil is calculated based on the percentage component fatty acids of the seed oil. The cetane number (CN), lower heating value (LHV) and higher heating value (HHV) of these fatty acid methyl esters (FAMEs) are empirically evaluated. The combustion characteristics /bio-diesel properties of Plectranthus mollis seed oil methyl esters (PMSOMEs) of Plectranthus mollis seed oil (PMSO) is compared with existing bio-diesels. The PMSO selected in this investigation convene the major specification of biodiesel standards. This work reports the suitability of PMSO for the bio-diesel productivity.*

### Graphical Abstract



Mechanism of the alkali-catalyzed transesterification of vegetable oils

**Keywords:** *Plectranthus mollis*, Industrial utilization, Biodiesel, Fatty acids, Fuel properties, *Plectranthus mollis* seed oil methyl esters.

## INTRODUCTION

Vegetable oil is basically triglyceride and has become popular due to some inherent advantages of being sustainable, biodegradable, non-toxic, eco-friendly, renewable, etc. The vegetable oil not only has industrial importance such as production of cosmetics, medicines, food, polymers, drugs, etc., researchers found that it acts as an alternative and sustainable energy source for diesel. A much more attractive loom that holds great potential is to take benefit of the great variety of fatty acids that are produced in wild plant species. This diversity of fatty acids represents a major opening for the balanced design of new technical oil qualities. These technical oil qualities are competent of satisfying demand from existing markets served by petrochemicals and can also lead to the development of new market applications due to unique chemical functionalities of seed oil fatty acids [**1-3**].

Alternative energy sources are the only solution to alleviate energy crisis due to fossil fuels depletion day by day, bio-based fuels are dragging much attention worldwide wherein, animal fats and vegetable oils are widely occurring lipids. We focused on the alternative liquid fuel to replace petro based fuel. Plant oils could only replace a small fraction of transport fuel divergent to this, the major problem posed by direct use of vegetable oil is its more viscosity compare to petrodiesel. The methods include dilution, micro emulsification, pyrolysis, catalytic cracking and transesterification [**4**] are some chemical technological routes developed to minimize the viscosity of vegetable or animal fat. The transesterification is sequence of conversion of triglycerides to diglycerides, and diglycerides to monoglycerides, offering glyceride conversion into glycerol yielding one ester molecule in each successive step. Transesterification is a most accepted technique in the biodiesel production. The by product like glycerol has tremendous industrial applications in cosmetic, pharma based, and other allied products. It has low viscosity and improved heating value which result in shorter ignition delay and longer combustion duration thus low particulate emissions. Biodiesel has become more attractive recently because of its eco-friendly nature. Its advantages include; its domestic origin, renewability, biodegradability, higher flash point, inbuilt lubricity and blending capability with petro diesel. The use of pure bio-diesel especially in the transport sector brings down the emissions of $CO_2$ by 80%. It has high cetane number than that of petro diesel. Biodiesel contains about 77% carbon, 12% hydrogen, and 11% oxygen by weight [**5**]. Biodiesel is a realistic fuel for future. It has more than 10% oxygen weight which reduces the carbon monoxide, oxides of sulphur and volatile organic content. Its flash point is more (423 K) compared to petro diesel (337 K). It is non-flammable hence non-explosive also. The mechanical problems associated with biodiesel are oxidative stability, poor low-temperature properties, and bit higher $NO_x$ exhaust emissions. The latter problem may be solved over time with the introduction of new exhaust emission control technologies [**6, 7**].

To reduce the processing and raw material cost of biodiesel it is recommended to use non- edible seed oils as raw materials, if unusual fatty acids are identified during analysis of seed oil, prior to transesterification process the isolation of industrially important unusual fatty acid/s to be carried out. Then, the rest of the material subjected for transesterification. Finally, the recovery of glycerol is important. Glycerol may be used as a chemical feedstock in the various products like polyurethanes, polyesters, polyethers, lubricants, wrapping and packaging materials, foods, drugs, cosmetics, tobacco products and products that displace existing petroleum derived materials. The manifestation of the biodiesel industry has generated a additional glycerol, which has initiated numerous efforts to find new products, and newer markets using glycerol as the resourceful chemical [**8-11**].

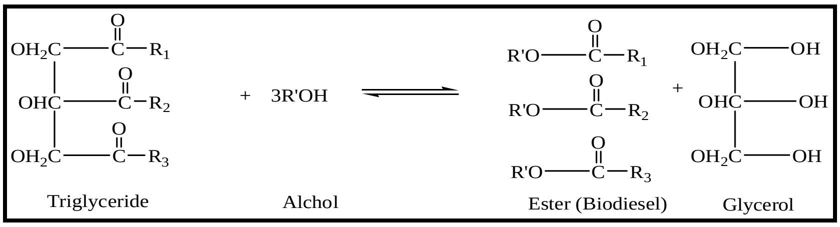
Biodiesel fuels obtained from different sources, varies with fatty acid profiles. Generally, it contains $C_{14}$–$C_{22}$ lower alkyl fatty acid esters. After transesterification FAMEs of vegetable oils have outstanding advantages over clean engine fuel alternatives. Currently, the commonly occurring fatty esters in biodiesel are those of palmitic acid, stearic acid, oleic acid, linoleic acid, and linolenic acid. Usually, soybean, palm, sunflower, safflower, cottonseed, rapeseed, and peanut oils are considered potential alternative fuels for diesel engines. The major seed oils sources like coconut, corn, cottonseed, canola, olive, peanut, safflower, soybean, and sunflower species are the multipurpose oils which are used for both biodiesel purpose as well as edible purpose. Many researchers from the past decades are proposing the growth of non-edible feedstocks like *Jatropha carcass, Calophyllum inophyllum, Moringa oleifera, Erica sativa gars, Croton megalocarpus, Cerberaodollam, Terminalia, Pongamia pinnata, Madhuca indica,* in forest and unused land for the commercialization of biodiesel [12-13]. Biodiesel can provide just such an interim solution. Knoth suggested that a mixture consisting primarily of methyl oleates of shorter chain esters derived from palmitoleic acid and esters derived from decanoic acid might be an ideal mixture. To meet this requirement transgenic crops are suggested [14-17].

The aim of this paper is to apply the already developed theoretical models substantiated with experimental results of fuel parameters on selected seed oil under study. The individual seed oil and its component fatty acid profile are utilized for evaluation of its combustion characteristics pertinent to the biodiesel profile. After the scrupulous investigation we have opted *Plectranthus mollis* seed oil fatty acid methyl esters for its combustion studies. *Plectranthus mollis* is an erect shrub or under-shrub. The leaves of this plant are used to stop bleeding, to cure fever and also as a mosquito repellent [18] Plectranthus of the family Lamiaceae is a large and widespread genus with a diversity of ethnobotanical uses. The genus Plectranthus, containing about 300 species, is found in Tropical Africa, Asia and Australia [19] The leaves are used as a vegetable [20] while roots are used to drive away evil spirits in India, Kenya and Tanzania [21] the seeds of *P. mollis* are fried in mustard oil and then massaged all over the body as an insect repellant.

## MATERIALS AND METHODS

**Oil extraction:** Soxhelet extraction method was used for the extraction of seed oil. The dried seeds of *P. mollis* were powdered and extracted with light petroleum to yield the oil. The analytical data of the oils were determined according to AOCS method [12]

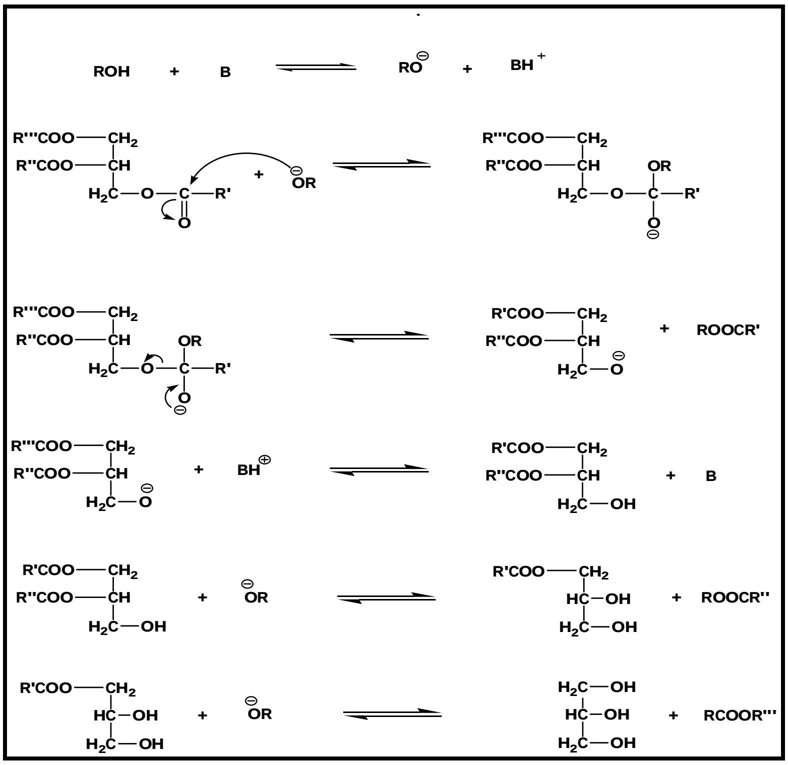
**Path of the Transesterification Reaction:** This is a reversible reaction. Sodium or potassium methanolate can be used for efficient conversion of fatty acids present in oils to their corresponding FAMES. The seed oil is treated in the ratio of 6:1 alcohol generally methanol or ethanol in presence of acid or base as catalyst and refluxed until completion of the esterification reaction. Then the mixture is transferred to separating funnel. It is allowed to stand for overnight. The lower layer containing glycerol, methanol and most of the catalysts was drained out. The upper layer containing methyl esters, some methanol, traces of the catalyst are cleaned thoroughly by washing with warm deionized water. Then, the residual methanol removed by rotary evaporation at around 70°C. Thus, obtained product containing FAMEs is used as biodiesel.




Path of the Transesterification reaction

**Table 1.** Physico Chemical Properties of *Plectranthus mollis* seed oil [12]

| | |
|---|---|
| Oil content in seeds (%) | 40.2 |
| Molecular weight of oil | 881.4 |
| Unsaponifiable matter (%) | 2.6 |
| Iodine value (g $I_2$ 100 $g^{-1}$ of oil) | 112.5 |
| Saponification value (mg KOH $g^{-1}$ of oil) | 196.5 |



Mechanism of the alkali-catalyzed transesterification of vegetable oils [21]

The selected seed oil is investigated for fuel properties as per Germany (DIN D6751) American (ASTM), and European (EN 14214) standards. The analytical data for this work such as % SFAs, %USFAs, IV, SN, CN, LHV, HHV are deployed in this work. The SN and IV were calculated empirically with the help of Eqs. (1) and (2) respectively or referred from the literature. Each data is correlated with one another. SN depends upon the molecular weight and the concentration of fatty acid esters i.e FAMEs of oil. However, IV, according to Eq. (2) unsaturated fatty acid components. Percent seed oil content SN, IV and percent fatty acid compositions of the selected seed oil is collected from the literature [12]

**Screening of combustion properties of P*lectranthus mollis* seed oil based on its component fatty acids or fames:** The selected seed oils were investigated for fuel properties as per American (ASTM), Germany (DIN D6751) and European (EN 14214) standards.

**Iodine value and Saponification number:** The standard mathematical models and the experimental data are compared. Theoretically, the Saponification value (SV) and Iodine value (IV) of fatty acid of this seed oil are calculated. The data obtained is used to establish suitability for biodiesel. Thus, SV and IV of seed oil are calculated using the equations (1) and (2) respectively based on compositions of fatty acid methyl ester [23] results obtained are very close to experimental values.

$$SV = \sum (560 \times Ai)/Mwi \qquad (1)$$

$$IV = \sum (254 \times D \times Ai)/Mwi \qquad (2)$$

Where, Ai is the percentage of component fatty acids, D is the number of double bonds and Mwi is the molecular mass of each component.

**Cetane number (CN):** The cetane number (CN) is an important parameter of the biodiesel/petro diesel fuel. Its prominence is useful during selection of Fatty acid profile / FAMEs to use as biodiesel.

The CN drives on how easily ignition occurs and the smoothness of the combustion. The CN affects a number of engine performance parameters like combustion, stability, drivability, white smoke, noise and emission of carbon monoxide and hydrocarbon. Generally, biodiesel has higher CN than conventional diesel fuel, which results in higher combustion efficiency. Using the equation (3) and is known closely match to the experimental values. The CN of FAMEs is calculated [24] using the mathematical model.

$$CN = 46.3 + (5458 / SV) – 0.225 x IV \qquad (3)$$

Importantly, FAMEs with higher CN are preferred which decides the heat content output and hence combustion quality. The CN and IV are inversely proportional. Literature reveals that, the increase of CN thereby IV decreases it means degree of unsaturation goes down leading to the solidification of FAMEs at high temperature. To overcome this problem, American biodiesel standard specified CN up to 65 [25]

**Higher heating value (HHV):** It is known that straight and processed vegetable oils used in diesel engines are the complex chemical mixture of fatty acid esters. The Higher heating value **(**HHV) of biodiesel is calculated using equation (4) [26].

$$HHV = 49.43 – (0.015 x IV) – (0.041 x SV) \qquad (4)$$

**Lower heating value (LHV):** The Lower heating value (LHV) of straight and processed vegetable oils is estimated with respect to equations (5) and (6) respectively based on bond energy of chemical structure of different fatty acid esters. The method predictive ability of LHV is more precise [27]
For FAMEs,

$$LHV = 0.0109(C/O)^3 - 0.3516(C/O)^2 + 4.2000(C/O) + 21.066 - 0.100 Ndb \qquad (5)$$

$$LHV = 0.0011(H/O)^3 - 0.0785(H/O)^2 + 2.0409(H/O) +20.992 - 0.100 Ndb \qquad (6)$$

Where, C = number of carbon atoms, H = number of hydrogen atoms, O= the number of oxygen atoms, and Ndb = number of double bonds.

**Cloud Point (CP) and Pour Point (PP):** The cold flow properties of biodiesel are also structure related properties and are determined by two parameters: cloud point (CP) and pour point (PP). CP is the temperature at which biodiesel becomes less transparent or hazy in appearance because of appearance of small crystals and is calculated. PP is the temperature where the biodiesel solidifies and stops flowing. This affects the performance of fuel lines, fuel pumps and injectors. Fatty acid esters with higher polyunsaturated esters yield lower cold flow properties whereas, monounsaturated esters yield moderate cold flow properties and higher saturated ester content yield higher cold flow properties. Cloud Point (CP) is the temperature at which biodiesel becomes less transparent or hazy in appearance because of appearance of small crystals. The CP and PP are calculated as per the equations (7) and (8) mentioned below [28].

$$CP = (0.526x C_{16})-4.992 \qquad (7)$$

$$PP = (0.571x C_{16})-12.24 \qquad (8)$$

**Flash Point (°C):** Flash point is of great significance to assess fire hazards or combustion consequences of liquid fuels and to control fire risks during storage, handling and transportation. It is defined as the minimum temperature at which the volatile fuel flashes or ignites for a moment when in contact with a flame or spark at a pressure of 101.325 kPa. Flash point of FAEs tend to increase with the increase in chain length and the degree of unsaturation, thus a higher flash point indicates a higher degree of stability and safety in handling the liquid fuel. It is calculated by using the following equation (9).

$$FP = 23.362\ N_c + 4.854\ N_{db} \qquad (9)\ [30]$$

Where, $N_c$ = weighted average number of carbon atoms and $N_{db}$ = weighted average number of double bonds.

Flash point of PMSOMEs was determined according to ASTM D92-12b standards. The empirically computed and experimentally determined values of flash point of BPME are 166.5°C and 190°C which are consistent with the values of existing biodiesel and higher than petro diesel (60°C-80°C).

## RESULTS AND DISCUSSION

More than 40% seed oil is recorded from *PMSO*. The major component fatty acids present are 46.5% linoleic acid 21.1% oleic acid, 13.6% stearic acid 7.2% palmitic acid. 6.1 % vernolic acid 3.2% sterculic acid and malvalic acid. . Apart from normal fatty acids mentioned above following are the unusual fatty acids. Those are 2.3% malvalic acid 3.2 % sterculic acid and 6.1 % vernolic acids. The physico chemical properties and component fatty acids in above said seed oil were shown in table 1 and table 2 respectively.

**Table 2.** Component fatty acids in *Plectranthus mollis* seed oil [**12**]

| Component fatty acids | | Mol. formula | Mol. Wt | % Fatty acid |
|---|---|---|---|---|
| **Name of the FA** | **No. of Carbons** | | | |
| Palmitic acid | 16:0 | $C_{16}H_{32}O_2$ | 256.43 | 7.2 |
| Stearic acid | 18:0 | $C_{18}H_{36}O_2$ | 284.48 | 13.6 |
| Oleic acid | 18:1 | $C_{18}H_{34}O_2$ | 282.42 | 21.1 |
| Linoleic acid | 18:2 | $C_{18}H_{32}O_2$ | 280.45 | 46.5 |
| Malvalic acid | 18:1 | $C_{18}H_{32}O_2$ | 280.45 | 2.3 |
| Vernolic acid | 18:1 | $C_{18}H_{32}O_3$ | 296.54 | 6.1 |
| Sterculic acid | 19:1 | $C_{19}H_{34}O_2$ | 294.48 | 3.2 |
| % TSFA | - | 20.8 | - | - |
| % TUSFA | - | 79.2 | - | - |

Where, TSFA= Total Saturated fatty acids, TUSFA= Total Unsaturated fatty acids

The molecular weight of PMSO is calculated based on component fatty acids. The MW of PMSO is 881.04 g $mol^{-1}$ which lies in the range of 855.64 –883.71 g $mol^{-1}$ respectively. Iodine value of this seed oil is 112.38 mg $KOH^{-1}$ not exceeding 120 which and best within the limit laid by EN 14214 for biodiesel. There is consistency in the SN. Generally, the FAMEs, which are mainly comprised of carbon chain lengths from 16 to 18, have boiling points in the range of 330–357°C, thus the specification value of 360°C is easily achieved. In the same context the FAMEs of PMSO meet the specification of 90/95% boiling point limit of 360°C specified in ASTMD6751 and in other biodiesel standards.

The CN of the PMSOMEs is observed to be 48.3. Biodiesel standards of USA (ASTMD 6751), Germany (DIN 51606) and European Organization (EN 14214) have set CN value as 47, 49 and 51, respectively [23]. The upper limit of CN (65) has been specified in US biodiesel standard (ASTMPS 121-99). Moreover, the CN of petro diesel is 42.6. Over all the empirically calculated CN value of PMSOMEs meet the requirement of biodiesel standards. This is depicted in fig.1 in the Combustion properties of biodiesel of PMSOMEs with existing biodiesels. There is very good trend in respect of combustion properties of biodiesel under study with the existing one. This infers that the PMSOMEs is a best candidate for the biodiesel production.

The LHV and HHV of the PMSOMEs are appreciable in acceptable range. The HHV of a fuel is a function of its C, H and O content. The LHV of straight and processed vegetable oils is estimated with as per equations (5) and (6) respectively. This is based on bond energy values of chemical structure of different fatty acids PMSOMEs fuel molecules. The empirically calculated results, for the PMSOMEs under this study indicates that there is a consistency of LHV that is with 39.5 MJ $Kg^{-1}$ in the species under investigation, this is quite lower than the LHV of petro diesel with 43 MJ $Kg^{-1}$. The European Biofuels Technology Platform 2011 reported the LHV for biodiesel as 37.1 MJ $Kg^{-1}$ [31]. The HHVs of biodiesel is relatively high. The HHVs of biodiesels for PMSOMEs is 39.5 MJ $kg^{-1}$ which is slightly lower than those of petro diesel 43 MJ $kg^{-1}$ or petroleum 42.0 MJ $kg^{-1}$, but are higher than coal 32–37 MJ $kg^{-1}$. Table 3 shows various combustion profiles of PMSOMEs.

**Table 3.** Combustion properties of Fatty acid methyl esters of seed oil of *Plectranthus mollis*

| S.No. | Fuel Parameters | PMSOME | SBSOME* | RPSOME* | SFSOME* | Petro-diesel* |
|---|---|---|---|---|---|---|
| 1. | Cetane Nunber | 48.3 | 45.0 | 54.4 | 49.0 | 42.0 |
| 2. | HHV (MJ $kg^{-1}$) | 39.55 | 39.7 | 37.0 | 40.5 | 42.5 |
| 3. | LHV (MJ $kg^{-1}$) | 37.86 | 33.50 | 32.80 | 33.5 | 43.1 |
| 4. | Cloud Point (°C) | -0.94 | -2.0 | –4.0 | 1.0 | –1 |
| 5. | Pour Point (°C) | -8.12 | -3.0 | –12 | NA | –16 |
| 6. | Flash Point (°C) | 169.56 | 178 | 170.0 | 183.0 | 45 |

* indicates some of the data obtained from, Ref. [32] NA=Not applicable
Where, SBSOME= Soya bean Seed Oil Methyl Esters RPSOME= Rape Seed Oil Methyl Esters
SFSOME=Sun Flower Seed Oil Methyl Esters.

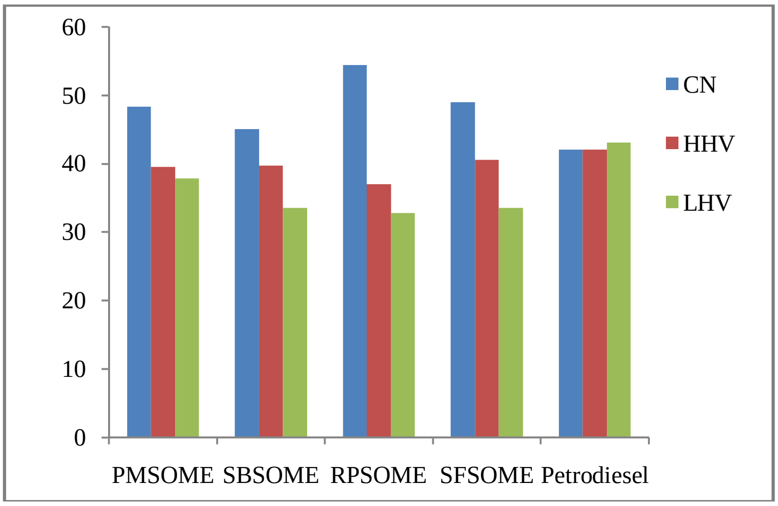



**Figure 1.** Combustion prosperities of PMSOMEs with existing biodiesels.

Where, CN= Cetane Number, HHV= Higher Heating Value, LHV= Lower Heating Value

The overall screening of PMSOMEs for biodiesel properties is comparable with the already existing biodiesels so also with the common petro diesel. This indicates that, the selected seed oil at this investigation can be a possible feed stock for the biodiesel production.

## APPLICATION

The PMSO is a promising feed stock for the production of biodiesel based on its component fatty acids and the combustion profile. The high saponification value of this seed oil is a useful data for the manufacture of various important industrially products such as shoe polish, liquid soap and shampoo products etc. The linoleic acid occur in cream which are essential in the human diet, infants grow poorly and develop skin lesion if fed a diet of non fat milk for prolonged period. The byproducts of the transesterification the purified glycerol can be applied in the food/cosmetic industries so also in the oleochemical based industry. Isolated unusual fatty acids can be utilized for possible industrial applications [**2, 32**].

## CONCLUSION

The FAMEs of PMSO meet the major specifications of US biodiesel standard (ASTMD 6751-02, ASTMPS 121-99), Germany (DIN V 51606) and European Standard Organization (EN 14214). The FAMEs of this seed oil of PM is found to be the most suitable for biodiesel production since meet the major specifications of biodiesel standards. However, still further research work is needed, to evaluate FAMEs for other property measures like, tribological studies, and long-term engine testing before full-fledged alternative fuel status. If many of such plants are grown in large scale on suitable wastelands, the bio-diesel produced from such seeds can supplement or replace the petro-diesel. Thus, these non edible seed oils are a highly capable source for bio-energy production.

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