



Thermodynamic Studies on Binary Blends of Palm and Melon Oils in Production of Biodiesel

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Environmental pollutions, energy problems and other uncertainties associated with the use of fossil fuel are some reasons which constitute the quest for eco-compatible fuel substitutes. Vegetable oils (VOs) are known raw materials that will remedy the situation. Palm oil (PO) and melon oil (MO) were extracted and blended in various proportions PO:MO (0:100, 10:90, 30:70, 40:60, 50:50, 60:40, 80:20, 100:0) for biodiesel (BDS) production through the process of trans esterification using a catalyst (sodium hydroxide, NaOH). Viscosities of the blends decrease as temperature increases. Experimental information was applied to Grunberg-Nissan (d-)parameter; the range of PO₅₀: MO₅₀ blend (-10.629 to -8.030) showed the least negative d-parameter values at all temperatures (283K to 323K) and viscosity deviations at various temperatures range between -20 to 21.

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Thermodynamic studies were carried out on the blends; the randomness of the system and negligible effect of inter- and intra- bonding energies of the system were defined by the increasing negative values of Gibbs free energy of mixing (ΔG^M) and zero enthalpies of mixing ($\Delta H^M = 0$). Energies of the produced BDS increase as calorific values of the blends increase (36 - 49 MJ/k) with an increase in the fractions of MO, and cloud point ranges from -1 to 0°C. The biodiesel properties were found to be comparable to the American Society for Testing and Material (ASTM) standard.

Keywords: Biodiesel; palm oil; melon oil; blends.

1. INTRODUCTION

Eco-friendly environment is the dream of the world today as the main sources of energy in the globe are petrochemical, hydroelectricity, coal and nuclear energy [1,2]. Petroleum-based fuel consumption for the past years contributed to the emission of greenhouse pollutants in the form of gases to our environment thus causing global warming of the earth's surface with increased emissions of combustion-generated gases [3,4,5]. Investigations have indicated that at the present rate of petroleum-based fuel utilization, there is fear of depletion following the nonrenewable nature of the natural resource [6,7,8]. Therefore, there is a need for an alternative type of fuel that can be both renewable and environmentally friendly. Using vegetable oil as a feedstock, biodiesel, therefore, is produced as alternative fuel which will serve as a good substitute for diesel fuel [9,10,11]. This potential energy source is renewable, and its use in biodiesel production will reduce environmental pollution and equally encourage more of its agricultural cultivation. Biodiesel is a liquid fuel obtained from oil-based biological material [12,13]. Edible oils from soybean, palm, and sunflower are mainly used for food purposes and have been predominantly used as sources of biodiesel owing to the increasing need for alternative sources of fuel [14,15]. Researchers have recommended that sources for diesel fuel can only be sustainable if the financial implication is reasonable, environmentally friendly, and always available. Because of this circumstance, vegetable oils are believed to meet such requirements; but the major drawback in using pure vegetable oils for biodiesel is the elevated viscosity, inadequate vaporization in the combustion chamber, difficulties of clogging of injectors, etc [16,17]. These disadvantages can be resolved if the vegetable oils are subjected to chemical modification through transesterification by mixing it with alkanol to get esters of the oil commonly called biodiesel with superior fuel characteristics [18,19]. Palm oil (PO) is an

edible vegetable oil extracted from the mesocarp of the fruit of the African oil palm, (*Elaeisguineensis*) [20,21]; and melon oil (MO) is extracted from melon seed (MES), a creeping oil-bearing crop which belongs to the family of *cucurbitaceae* [22,23].

In this paper, we report the work of extracting MO and PO, and blends of both oils in various proportions PO:MO (0:100, 10:90, 30:70, 40:60, 50:50, 60:40, 80:20); both oils and their blends were used in the production of biodiesel through transesterification method using sodium hydroxide (NaOH) as a catalyst. The produced biodiesel using different proportions of PO and MO was characterized and compared with the ASTM.

2. METHODOLOGY

2.1 Sample Collection and Preparation

2.1.1 Palm fruit collection and extraction of PO

The palm fruit bunch was harvested from a palm tree on a farm at Umuahunwo Village, Aluu in Ikwerre Local Government Area of River State. The stalk-carrying section of the palm fruits was cut out from the bunch, sterilized, and divided into seven portions. Oils from each portion were mechanically extracted for seven days (one portion for each day).

2.1.2 Melon seeds collection and extraction of MO

Non-dehusked melon seeds were purchased from "boundary" market Umuahunwo Village, Aluu in Ikwerre Local Government Area of River State; then de-husked, ground, and were divided into seven portions. Extraction of oil in each portion was carried out each day for seven days, one portion for one day using Soxhlet extraction. The oil in the seeds of melon was extracted through continuous extraction in Soxhlet

apparatus by loading 70 g of ground MES in the thimble of the extractor and 700 ml of n-hexane was dispensed into the 1000 ml round bottom flask together with some pieces of anti-bomb material. The round bottom flask kept in a water bath was attached to the thimble connected to the condenser. The condenser was in contact with the essential water pipes so that water can flow in and out of the condenser to ensure adequate cooling of the n-hexane vapours as the water bath is switched on. The Process was completed when the ground MES in the thimble turned colourless. The residue was taken out of the thimble, weighed and replaced with a new sample. The extracted oil was recovered from the n-hexane extract mixture using a rotary evaporator and labeled properly. The above extraction method was repeated for the other portions.

2.2 Blending of PO and MO

The PO and MO were mixed in various proportions (0:100, 10:90, 30:70, 40:60, 50:50, 60:40, 70:30, 80:20, 100:0) and labeled accordingly: PO₀:MO₁₀₀, PO₁₀:MO₉₀, PO₃₀:MO₇₀, PO₄₀:MO₆₀, PO₅₀:MO₅₀, PO₆₀:MO₄₀, PO₈₀:MO₂₀ and PO₁₀₀:MO₀.

2.3 Viscosity

Kinematic viscosities of the oils and their mixtures were determined at various temperatures (30-70°C) using a Ubbelohde viscometer. Then the corresponding dynamic viscosities were determined using the equation

$$\eta = \nu\rho = ktp \quad (1)$$

Where,

η = dynamic viscosity of PO/MO/blends (Kgm⁻¹s⁻¹)

ν = kinematic viscosity (mm²s⁻¹)

t = time (s)

k = viscometer constant (m²s⁻²)

ρ = the density of PO or MO or blends (Kgm⁻³)

2.4 Density

Densities of the PO, MO and their blends were obtained using the equation

$$\rho = \frac{m}{v} \quad (2)$$

Where,

ρ = the density of (PO or MO or blends).

m = the mass of (PO or MO or blends)

v = the volume of (PO or MO or blends)

2.5 Data Analysis

Experimental results obtained were applied to the following models:

2.5.1 Viscosity deviation ($\Delta\eta$)

Deviation in viscosities is calculated with the expression in the equation:

$$\Delta\eta = \eta_m - (x_1\eta_1 + x_2\eta_2) \quad (3)$$

Where:

η_m = viscosity PO and MO blend

x_1 and x_2 = mole fractions of PO and MO components respectively.

η_1 and η_2 = viscosities of PO and MO components respectively.

2.5.2 d- parameter

$$d = \ln\eta - \frac{X_1\ln\eta_1 + X_2\ln\eta_2}{X_1X_2} \quad (4)$$

Where,

x_1 = mole fraction of PO

x_2 = mole fraction of MO

η = viscosity of mixture (Kgm⁻¹s⁻¹)

η_1 = viscosity of PO

η_2 = viscosity of MO

2.6 Thermodynamic Studies

Studies were reformed by ascertaining the thermodynamic properties, Gibb's free energy of mixing (ΔG^M), entropy of mixing (ΔS^M) and enthalpy of mixing (ΔH^M) at equilibrium using equations 5, 6, 7 respectively as follows:

$$\Delta G^M = nRT (X_1\ln X_1 + X_2\ln X_2) \quad (5)$$

$$\Delta S^M = -nR(X_1\ln X_1 + X_2\ln X_2) \quad (6)$$

$$\Delta H^M = \Delta G^M + T\Delta S^M \quad (7)$$

Where,

x_1 = mole fraction of PO

x_2 = mole fraction of MO

n = number of moles (mol)

R = gas constant (JK⁻¹mol⁻¹)

T = temperature (K)

2.7 Biodiesel Production

Solution of sodium methoxide was prepared by dissolving 0.88g of NaOH in 10 ml of methanol in a conical flask with a stopper and shaking for 1 hour to obtain CH₃ONa solution which was mixed with 50 ml of the oil (PO / MO/blends); and with continuous stirring using a magnetic stirrer at 60°C for 20 min. The mixture was subsequently

introduced into a separatory funnel and kept for 24 hours. The BDS produced was washed with distilled water by carefully dropping 40 ml of distilled water down the side of the glass. After washing, the sample was kept for 30 min before the aqueous layer was drained off; the remaining trace of water was gently evaporated at 70 °C [24].

3. RESULTS AND DISCUSSION

3.1 Data Presentation

Table 1. Kinematic viscosities of palm oil, melon oil and their mixtures

Binary blends of PO and MO	Kinematic viscosity (mm ² /s)				
	30°C	40°C	50°C	60°C	70°C
PO ₀ :MO ₁₀₀	25.43	22.75	19.81	15.02	12.05
PO ₁₀ :MO ₉₀	30.97	27.03	24.59	15.96	12.80
PO ₃₀ :MO ₇₀	35.43	31.91	29.05	16.88	13.54
PO ₄₀ :MO ₆₀	33.40	31.33	28.87	16.85	14.34
PO ₅₀ :MO ₅₀	41.40	36.70	34.11	25.15	22.20
PO ₆₀ :MO ₄₀	36.40	31.58	29.79	21.85	19.39
PO ₈₀ :MO ₂₀	44.44	42.11	40.55	36.11	34.15
PO ₁₀₀ :MO ₀	57.40	46.94	43.80	25.69	20.77

Table 2. Densities of palm oil, melon oil and their mixtures

Binary blends of PO and MO	Density (g/m ³)				
	30 °C	40 °C	50 °C	60 °C	70 °C
PO ₀ :MO ₁₀₀	0.910	0.905	0.891	0.887	0.885
PO ₁₀ :MO ₉₀	0.912	0.906	0.893	0.888	0.886
PO ₃₀ :MO ₇₀	0.914	0.907	0.895	0.889	0.887
PO ₄₀ :MO ₆₀	0.913	0.906	0.894	0.889	0.887
PO ₅₀ :MO ₅₀	0.915	0.909	0.895	0.888	0.887
PO ₆₀ :MO ₄₀	0.916	0.911	0.895	0.889	0.887
PO ₈₀ :MO ₂₀	0.920	0.917	0.897	0.889	0.888
PO ₁₀₀ :MO ₀	0.925	0.918	0.902	0.890	0.889

Table 3. parameters from viscosity deviation model

Binary blends	Viscosity deviation (Δη)				
	Temperature				
	30°C	40°C	50°C	60°C	70°C
PO ₁₀ :MO ₉₀	-20.22	-15.22	-14.56	-7.66	-6.28
PO ₃₀ :MO ₇₀	-9.00	-5.74	-5.54	-4.71	-3.88
PO ₄₀ :MO ₆₀	-8.40	-3.85	-3.25	-2.20	-1.18
PO ₅₀ :MO ₅₀	2.77	3.96	4.39	5.72	6.55
PO ₆₀ :MO ₄₀	0.96	1.28	2.50	3.51	4.62
PO ₈₀ :MO ₂₀	15.37	16.60	18.01	19.85	21.11

Table 4. d- parameter and thermodynamics for various blends of palm oil and melon oil

Temperature/K	Parameter	PO ₁₀ :MO ₉₀	PO ₃₀ :MO ₇₀	PO ₄₀ :MO ₆₀	PO ₅₀ :MO ₅₀	PO ₆₀ :MO ₄₀	PO ₈₀ :MO ₂₀
303	D	-28.623	-12.338	-10.934	-10.629	-12.00	-20.585
	$\Delta G^M/kJmol^{-1}$	-15.642	-27.211	-29.888	-30.695	-30.043	-22.321
	$\Delta S^M/JK^{-1}mol^{-1}$	1.72E-4	2.97E-4	3.33E-4	3.33E-4	3.27E-4	2.44E-4
	$\Delta H^M/kJmol^{-1}$	0	0	0	0	0	0
313	D	-27.584	-11.800	-10.389	-10.117	-11.351	-19.463
	$\Delta G^M/kJmol^{-1}$	-16.158	-28.109	-30.874	-31.709	-31.034	-23.058
	$\Delta S^M/JK^{-1}mol^{-1}$	1.66E-4	2.88E-4	3.16E-4	3.23E-4	3.16E-4	2.36E-4
	$\Delta H^M/kJmol^{-1}$	0	0	0	0	0	0
323	D	-26.244	-11.297	-9.963	-9.772	-10.95	-19.280
	$\Delta G^M/kJmol^{-1}$	-16.674	-29.007	-31.861	-32.722	-32.026	-23.794
	$\Delta S^M/JK^{-1}mol^{-1}$	1.61E-4	2.79E-4	3.07E-4	3.13E-4	3.07E-4	2.29E-4
	$\Delta H^M/kJmol^{-1}$	0	0	0	0	0	0
333	D	-23.264	-10.264	-8.892	-8.692	-9.736	-16.525
	$\Delta G^M/kJmol^{-1}$	-17.191	-29.905	-32.847	-33.735	-33.017	-24.531
	$\Delta S^M/JK^{-1}mol^{-1}$	1.56E-4	2.70E-4	2.97E-4	3.03E-4	2.97E-4	2.22E-4
	$\Delta H^M/kJmol^{-1}$	0	0	0	0	0	0
343	D	-10.575	-9.499	-8.156	-8.030	-9.039	-11.411
	$\Delta G^M/kJmol^{-1}$	-17.707	-30.803	-33.834	-34.748	-34.009	-25.268
	$\Delta S^M/JK^{-1}mol^{-1}$	1.52E-4	2.62E-4	2.89E-4	2.94E-4	2.89E-4	2.16E-4
	$\Delta H^M/kJmol^{-1}$	0	0	0	0	0	0

Table 5. Characterization of biodiesel produced from PO and MO and their blends

Parameter	PO ₀ :MO ₁₀₀	PO ₁₀ :MO ₉₀	PO ₃₀ :MO ₇₀	PO ₄₀ :MO ₆₀	PO ₅₀ :MO ₅₀	PO ₆₀ :MO ₄₀	PO ₈₀ :MO ₂₀	PO ₁₀₀ :MO ₀	ASTM
Specific gravity at 40 °C	0.876	0.884	0.885	0.886	0.889	0.890	0.894	0.899	0.860 - 0.900 (D445)
Kinematic Viscosity/mm ² s ⁻¹	4.09	5.38	5.52	5.60	5.69	5.74	6.03	6.21	1.90 - 6.00 (D445)
API gravity	37.28	33.41	32.75	31.64	30.81	30.19	29.15	28.46	
Flash Point/ °C	135	128	115	100	99	98	93	90	90 – 130 (D93)
Cloud Point/ °C	-1	-1	-1	-1	-1	-1	0	0	-15 – 5 (D2500)
Aniline Point/ °C	75	71	69	68	65	63	61	58	
Density at 30 °C /g/m ³	0.876	0.884	0.885	0.886	0.889	0.890	0.894	0.899	0.86 – 0.89 (D 1298)
Cetane no.	58	55	55	53	51	50	49	45	47 – 67 (D664)
Carbon residue/%	0.031	0.029	0.028	0.027	0.026	0.025	0.024	0.023	0.015 – 0.050 (D5453)
Calorific value/MJ/k	49	45	43	42	39	38	36	31	38 – 43 EN 14213

API= American Petroleum Institute

3.2 Discussion

The viscosity and density of PO, MO and their mixtures (Tables 1 and 2) decreased with an increase in temperature. This suggests that temperature has a noticeable effect on the viscosities and densities of PO, MO and their blends. Viscosity and density are essential characteristics of oil quality and they respond to variations in temperature, concentration and structure [25,26].

3.2.1 Viscosity deviation

The negative and positive values of viscosity deviation (Table 3) indicate the presence of weak and strong intermolecular forces of attraction respectively upon mixing PO and MO. Thus, viscosity deviation interprets the collision pattern of the molecules of PO, MO and their blends with temperature. A decrease in negative values ($\text{PO}_{10}:\text{MO}_{90} - \text{PO}_{40}:\text{MO}_{60}$) and an increase in positive values ($\text{PO}_{50}:\text{MO}_{50} - \text{PO}_{80}:\text{MO}_{20}$) indicate an increase in the frequency of collisions between molecules of PO and MO in the blends as the temperature increases [27,28].

3.2.2 Grunberg-Nissan (d-parameter)

The viscosity of the binary component mixture is linked to its mole fraction by an expression derived by Grunberg-Nissan (d-parameter). It gives the extent of interaction among the molecular constituents of a binary mixture at a specified temperature [29,30]. d-parameter deductions were done by the application of measured dynamic viscosity values. The result in Table 4 shows a rise in the d-parameter with the increase in mole fraction in some cases, signifying stronger interaction of molecules at increased mole component of PO in the various blends ($\text{PO}_{10} - \text{PO}_{50}$), and thereafter, decrease with further increase in PO mole fractions ($\text{PO}_{60} - \text{PO}_{80}$).

3.2.3 Thermodynamics of mixing

When solids, liquids or gases combine, the thermodynamic quantities of the system experience a change because of the mixing. Focusing specifically on the effect of such a combination on Gibbs free energy (ΔG^M), entropy (ΔS^M) and enthalpy (ΔH^M) of mixing PO and MO; ΔG^M determines if the process will be spontaneous, ΔS^M specifies the extent of the randomness of the system, while ΔH^M is the resultant effect on absorbed or released heat by

the interactions between PO and MO [31,32]. The tendency of PO and MO to mix is indicated in the Gibbs free energy change of mixing presented in Table 4. The negative values of ΔG^M signify the presence of sufficient free energy within the system, which is discharged during mixing, such that the molecules of both liquids exert energy as they interact, which caused the increase in the disorderliness of the system and its surrounding. The amount of ΔG^M increased in negativity with the increase in mole fractions of PO_{10} to PO_{50} and decreases upon further increase in P_{60} to P_{80} fractions. The higher negativity of ΔG^M values was found with PO_{10} to PO_{50} at all experimental temperatures, indicating the greatest tendency of a molecular blend, which rose with temperature. Changes in the values of ΔG^M with mole ratios of PO and MO within the experimental temperature substantiate the dependence of ΔG^M on the temperature and value of the constituents of the blend. Therefore, if the free energy change involved with the formation of a mixture at particular pressure and temperature is greater than that of the component liquids, then the process will be non-spontaneous; on the other hand, the process becomes spontaneous if the free energy value of the product is lesser than those of the component liquids [33,34]. Furthermore, the favourability of a reaction depends on the overall free energy, ΔG^M of reactants and products; and the changing pattern of randomness (ΔS^M) of the system [35]. Throughout this experiment, values of ΔH^M is zero reflecting the negligible effect of intra and inter-bonding forces at all temperatures, and the dominance of solvent-solvent interaction; while the positive values of (ΔS^M) suggest a reasonable level of randomness and that the molecules dispersed equally in the binary system.

3.2.4 Biodiesel characterization

The specific gravity of biodiesel is the proportion of the density of biodiesel to that of water at a specific temperature and pressure [36]. It is a field test that dictates impurities in biodiesel. It equally determines how long a volume of biodiesel can last in an engine, and also signifies the energy level of biodiesel. An increase in the specific gravity of biodiesel implies an increase in energy, and the ASTM range for specific gravity is 0.86 – 0.90. The entire blend in Table 5 shows an approximate worth of 0.90. Density and specific gravity values of the biodiesel of PO, MO and their combinations shown in Table 5 increased with an increase in the fractions of PO indicating that PO is more viscous than MO.

The viscosity of BDS is a measure of its opposition to flow, and BDS with a high level of viscosity can hinder the efficiency of engine operation [37,38]. Viscosity levels in the entire blends are reduced as MO fractions are increased. This suggests that increasing MO fractions ($MO_{40} - MO_{100}$) in the BDS encouraged adequate combustion and will reduce exhaust smoke; while BDS from reduced MO fraction ($MO_{20} - MO_0$) is not ideal in cold weather, because of higher viscosity levels since the decrease in temperature causes an increase in viscosity [39,40].

Calorific value of biodiesel is the quantity of heat produced on its complete combustion, and it signifies the usable energy in the biodiesel. Calorific Values of PO and MO in this research increase with increase in the fractions of MO in the blends, and this suggests higher energy in MO than PO, because the viscous nature of PO offers more resistance and therefore slows down the combustion process in PO than MO; hence, lower energy is produced out of molecules of PO than MO.

Cetane number indicates how much a given BDS can burn in an engine [41,42]. The ASTM recommended range for biodiesel is 47 – 67 min (Table 5). As the cetane number increases, the ignition delay decreases and combustion increases [43,44]. It was noted in this research that the cetane number rises with the increase in the fractions of MO, indicating that molecules of MO will burn more easily in the engines; and therefore possess more combustion qualities than PO.

Flash point of BDS is the temperature at which its vapour is ignited. It encourages safety decisions in the proper handling and storing of BDS [45] the flash point values for PO, MO and their blends are presented in Table 5. All flash point values obtained in the experiment were higher than 90°C, and therefore, safe in terms of storage, transportation and fire hazard; since biodiesel with a flash point above 66°C is considered safe [46].

Cloud point of BDS is the temperature at which solid crystals are formed in the BDS [47]. The cloud point values for BDS obtained from raw PO and MO, as well as their blends (Table 5) were within the ASTM range (-15.0 – 5.0°C). The cloud point values obtained for most of the blends were (-1°C) except those of $PO_{80}:MO_{20}$ and $PO_{100}:MO_0$ with values of 0°C.

This could be because PO is more saturated than the MO, as saturation favours a high cloud point, and therefore leads to crystallization; indicating that the produced biodiesel will burn successfully in an engine even at low temperature.

Carbon residue indicates the tendency of biodiesel to deposit carbon on the engines causing various operational problems like corrosion and blockage of the nozzle [48]. The carbon residue (Table 5) is within the accepted ASTM limit of 0.015 – 0.050 %. The lower value of carbon residue in this research shows that the produced biodiesel can last long in storage without the problem of carbon deposition in the environment, especially when the fractions of PO is increased ($PO_{10}:MO_{90} - PO_{90}:MO_{10}$) in the blends.

American petroleum institute (API) gravity varies inversely to specific gravity, such that API gravity increases as the energy of biodiesel decreases [49]. $PO_0:MO_{100}$ and $PO_{100}:MO_0$ have the highest and lowest API values of 37.28 and 28.46 respectively; while values of the various blends were decreasing with the rise in the fraction of PO, suggesting that the energy of the produced biodiesel increased with an increase in the fractions of MO.

Aniline point (AP) is the temperature at which equal volumes of aniline ($C_6H_5NH_2$) and lubricant oil are miscible [50]. The amount of aromatics in BDS increases as the AP decreases and a low AP is a reflection of higher aromatics, while a high AP reflects lower aromatics content [51]. The highest AP value in the experiment (75°C) was recorded in $PO_0:MO_{100}$, while the lowest value (58°C) was recorded in $PO_{100}:MO_0$. AP values decreased with increase in the fraction of PO in all mixtures, which suggests an increase in the aromatic content of the produced BDS as molecules of PO is increased in the blends; thereby raising the ignition features of the biodiesel.

4. CONCLUSION

On the whole assessment, the $PO_{50}:MO_{50}$ blend at 70°C exhibited the lowest value of ΔG^M , signifying the highest favourable interaction with relatively little energy for complete burning of the BDS in the engine. The ΔH^M values were zero, suggesting non-interference of ion pair formation in the entire PO:MO blends; while the values of the ΔS^M which are between 1.52×10^{-4} and

$3.33 \times 10^{-4} \text{ JK}^{-1} \text{ mol}^{-1}$ suggest minimal randomness of molecules within the mixtures of PO and MO. It was observed that the biodiesel produced does not require a very high operating temperature as the cloud point ranges from -1 to 0°C. The features of the produced biodiesel from result characterizations fall within the specification of ASTM for biodiesel. Hence, palm oil and melon oil are considered good feedstock for biodiesel production.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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