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Physicochemical Properties of Jatropha Curcas Biodiesel + Diesel Fuel No. 2 Binary Mixture at $T = (288.15 \text{ to } 308.15)$ K and Atmospheric Pressure

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ABSTRACT: Environmental issues, the growing demand for energy, political concerns, increasing crude oil prices and the medium-term depletion of petroleum created the need for the development of vegetable oils as alternative fuels. Vegetable oil-based fuels (bio fuels) are promising alternative fuels for diesel engines because of their environmental and strategic advantages. To design equipment for biofuel production and an optimizing process for biodiesel production, their thermophysical properties must be known. In this work, the *Jatropha curcas* biodiesel was prepared, and thermophysical properties, densities (ρ_{12}), and speed of sound (u_{12}) for *J. curcas* biodiesel (1) + diesel fuel no. 2 (2) binary mixtures were measured as functions of composition at temperatures ranging from $T = (288.15 \text{ to } 308.15)$ K and atmospheric pressure. The observed data have been utilized to evaluate the excess molar volume, V_{12}^{E}, of this binary mixture. This binary mixture (blend) exhibits a temperature-dependent behavior, and densities decrease linearly with temperature.

■ INTRODUCTION

In recent years systematic efforts have been made by several research workers[1−4] to use vegetable oils as fuel in engines. Vegetable oil-based fuels are sustainable sources of fuel as their burning produce less emissions (CO_2, SO_x, NO_x, etc). These oils can be used as fuels in conventional diesel engines directly without any modification. However, diesel engines with vegetable oils suffer from operational and durability problems for long-term operation as vegetable oils are much more viscous, relatively more reactive to oxygen, and have higher cloud-point and pour-point temperatures than petroleum-based diesel fuel. These problems can be solved if the vegetable oils are chemically modified to biodiesel, which is similar in characteristics to diesel.[5] Biodiesel has substantially different properties than vegetable oils and results in better engine performance. However, most of the vegetable oils used for biodiesel production are also used in food industry. This fact creates market conflict between biodiesel and edible oils, leading to even greater biodiesel costs.[6] Great attempts have been made to produce biodiesel from nonedible oils such as *Jatropha curcas* oil. Because of the presence of phorbol ester in its composition, *J. curcas* oil is not suitable for food and feed applications.[7] Therefore, interest in using curcas oil as a feed stock for biodiesel production has rapidly grown in recent years.[8−15] To overcome the higher biodiesel costs blending biodiesel with diesel fuel is one of the solutions. Therefore, it is important to know that the basic properties of blends and rheological properties of fuel must be studied to determine behavioral and predictive information for the design and optimization of heating and fuel injection systems.[16] Density data are important in numerous chemical engineering unit operations. Fuel density data as a function of temperature are needed to model the combustion processes and other applications. Many studies have been done involving blending of vegetable oils or biodiesel with diesel fuel, recently.[17−24] However, binary mixtures (blends) of biodiesel and diesel fuel were rarely studied and characterized. This work is motivated by the fact that there is no comprehensive study of biodiesel with diesel fuel. The specific objective of this work is to prepare binary mixture (blend) of *J. curcas* biodiesel and diesel fuel (no. 2) and to measure the densities (ρ) and speed of sound (u) as a function of composition at several temperatures (288.15, 293.15, 298.15, 303.15, and 308.15) K and atmospheric pressure.

■ EXPERIMENTAL SECTION

Materials. Commercially available automotive diesel fuel no. 2 was used in this study. Petroleum-derived diesel is composed of about $w = 0.75$ saturated hydrocarbons (primarily paraffins including *n*, *iso*, and cycloparaffins) and $w = 0.25$ aromatic hydrocarbons (including naphthalenes and alkylbenzenes) as reported in Table 1. The average chemical formula for common diesel fuel is $C_{12}H_{23}$, ranging approximately from $C_{10}H_{20}$ to $C_{15}H_{28}$.[25,26]

Methanol (mass fraction $w = 0.99$, Sigma-Aldrich) and pure H_2SO_4 (mass fraction $w = 0.98$, Merck) were used as a catalyst for transesterification.

Biodiesel Preparation. *J. curcas* seeds were collected, and kernels were separated from the shells for oil extraction. The kernels were then cold-pressed, and approximately 278 g of oil was recovered from 1000 g of kernels for duplicate samples (27.8 % oil content). Production of biodiesel was carried out in a batch reactor through the transesterification of *J. curcas* oil. The reactor included a 2.5 L jacketed glass, mechanical stirrer (Kika Werke)

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fitted with a stainless steel propeller, thermometer, and water-cooled reflux condenser. To prepare and control reaction temperature, a RCS and RC6 (LAUDA) hot water circulation bath was employed. The reactor was filled with *J. curcas* oil and acid-catalyzed transesterification with H_2SO_4 in methanol was used to produce fatty acid methyl ester (FAME). After the end of transesterification, the glycerol was separated by a separatory funnel. The produced methyl esters were washed with hot 0.1 % aqueous tannic acid solution three times. The excess water and methanol were removed on a rotary evaporator at atmospheric pressure.

Fatty Acid Composition Analysis. The composition of the *J. curcas* biodiesel was analyzed by gas chromatography (GC) with flame ionization detection (FID) using a 50 % cyanopropyl polysiloxane phase (Agilent Technologies, DB-23; 30 m × 0.25 mm inner diameter). Helium was used as the carrier gas, and the gas line was equipped with oxygen. *J. curcas* biodiesel was found to contain oleic acid, linoleic acid, and palmitic and stearic acid (reported in Table 2). Palmitic and stearic acid are the major saturated fatty acids found in *J. curcas* biodiesel. It contains approximately 80 % unsaturated fatty acids. From this composition the molecular weight of jatropha biodiesel was calculated to be 870 $g \cdot mol^{-1}$. Further, the average molecular weight of diesel fuel no. 2 was calculated to be 233 $g \cdot mol^{-1}$. However the average molecular weight of diesel fuel no. 2 has been reported as (200 to 230) $g \cdot mol^{-1}$).[27,28] Reason being diesel fuel no. 2, like other petroleum products, is not specific chemical compound but a complex mixture of organic compounds separated into ranges by their boiling point.

Apparatus and Procedure. The binary system was prepared by mass using a Mettler mass balance (Switzerland, model AE-200) with an accuracy of ± 0.0001 g. The more volatile component was filled directly into the airtight Stoppard 5 cm^3 glass vial and then weighed. The second component was then injected into the vial through the stopper by means of a syringe. This method prohibited significant evaporation and contamination, which would have resulted in composition errors. The possible error in mole fraction using this procedure is estimated to be lower than 0.001. Densities (ρ) and speed of sound (u) of pure components and binary mixture (blend) were measured using an Anton Paar digital vibrating glass tube densimeter (model DSA 5000) at (288.15, 293.15, 298.15, 303.15, and 308.15) K in the manner described elsewhere.[29,30] The densities of the pure components together with their literature[31−33] values are given in Table 3. The difference in the measured and literatures values for diesel fuel no. 2 may be due to irreproducible composition of pure component. The uncertainty in density measurement is $2 \cdot 10^{-3}$ $kg \cdot m^{-3}$ and for speed of sound is ± 0.1 $m \cdot s^{-1}$.

Table 1. Composition of Diesel Fuel No. 2[25,26]

	volume %		
carbon no.	paraffins	cycloparaffins	aromatics
C10	0.9	0.6	0.4
C11	2.3	1.7	1.0
C12	3.8	2.8	1.6
C13	6.4	4.8	2.8
C14	8.8	6.6	3.8
C15	7.4	5.5	3.2
C16	5.8	4.4	2.5
C17	5.5	4.1	2.4
C18	4.3	3.2	1.8
C19	0.7	0.6	0.3

RESULTS AND DISCUSSION

J. curcas biodiesel was prepared via acid-catalyzed tranesterification and analyzed by gas chromatography. The composition of both *J. curcas* biodiesel and commercially available diesel fuel no. 2 has been reported in Tables 1 and 2, respectively.

Densities (ρ_{12}) and speed of sound (u_{12}) data of *J. curcas* biodiesel (1) + diesel fuel (2) binary mixture (blend) were measured as a function of composition at temperatures ranging from T = (288.15 to 308.15) K. The excess molar volume, V_{12}^E, were calculated from density data using eq 1.

$$V_{12}^{E} = (x_1 M_1 + x_2 M_2)/\rho_{\text{mix}} - (x_1 M_1/\rho_1) - (x_2 M_2/\rho_2) \quad (1)$$

Table 3. Densities (ρ) and Speed of Sound (u) Data for Pure Diesel Fuel No. 2 and *J. curcas* Biodiesel at Temperatures Ranging from T = (288.15 to 308.15) K and Atmospheric Pressure

		ρ		u
	T	$g \cdot cm^{-3}$		$m \cdot s^{-1}$
component	K	expt.	lit.	expt.
J. curcas biodiesel	288.15	0.884141	0.8809[31]	1429.43
	293.15	0.880501		1411.28
	298.15	0.876862		1393.41
	303.15	0.873227		1375.60
	308.15	0.869588	0.8650[32]	1357.91
diesel fuel no. 2	288.15	0.824386		1377.48
	293.15	0.820996		1359.60
	298.15	0.817494		1340.79
	303.15	0.813986	0.8500[33]	1322.14
	308.15	0.810476	0.8410[32]	1303.63

Table 2. Fatty Acid Composition of *J. curcas* Biodiesel

fatty acid mass percent		
structure	common name	composition (mass %)
$CH_3\ (CH_2)_{14}COOH$	16:0 (palmitic)	15.88
$CH_3\ (CH_2)_{16}COOH$	18:0 (stearic)	6.15
$CH_3\ (CH_2)_7CHCH\ (CH_2)_7COOH$	18:1 (oleic)	41.85
$CH_3(CH_2)_4CHCHCH_2CHCH(CH_2)_7COOH$	18:2 (linoleic)	35.32

Table 4. Densities (ρ_{12}), Speed of Sound (u_{12}), and Excess Molar Volumes, V_{12}^{E} for *J. curcas* Biodiesel (1) + Diesel Fuel No. 2 (2) at Temperatures Ranging from T = (288.15 to 308.15) K and Atmospheric Pressure

	ρ_{12}	V_{12}^{E}	u_{12}
x_1	$g \cdot cm^{-3}$	$cm^3 \cdot mol^{-1}$	$m \cdot s^{-1}$
T = 288.15 K			
0.0000	0.824386	0.0000	1377.48
0.0443	0.832586	0.0389	1383.50
0.1134	0.842555	0.1039	1391.28
0.1775	0.849706	0.1511	1397.26
0.2062	0.852436	0.1640	1399.61
0.2457	0.855816	0.1724	1402.54
0.2868	0.858948	0.1664	1405.29
0.3202	0.86125	0.1505	1407.36
0.3721	0.864467	0.1047	1410.27
0.4104	0.866602	0.0553	1412.25
0.4628	0.869246	−0.0322	1414.81
0.5233	0.871956	−0.1546	1417.42
0.5674	0.873732	−0.2539	1419.10
0.6326	0.876082	−0.4013	1421.34
0.6872	0.87782	−0.5123	1423.01
0.7436	0.87941	−0.5978	1424.51
0.8251	0.881355	−0.6288	1426.38
0.8672	0.882199	−0.5813	1427.25
0.9252	0.883183	−0.4199	1428.35
1.0000	0.884141	0.0000	1429.43
T = 293.15 K			
0.0000	0.820996	0.0000	1359.60
0.0443	0.82912	0.0549	1365.44
0.1134	0.839022	0.1334	1373.07
0.1775	0.846141	0.1851	1378.91
0.2062	0.848863	0.1979	1381.21
0.2457	0.852236	0.2046	1384.14
0.2868	0.855363	0.1958	1386.86
0.3202	0.857663	0.1765	1388.90
0.3721	0.860877	0.1257	1391.89
0.4104	0.863011	0.0717	1394.21
0.4628	0.865652	−0.0208	1396.55
0.5233	0.868358	−0.1484	1399.18
0.5674	0.870129	−0.2497	1401.07
0.6326	0.872472	−0.3997	1403.58
0.6872	0.874203	−0.5113	1405.29
0.7436	0.875785	−0.5958	1406.74
0.8251	0.87772	−0.6248	1408.57
0.8672	0.87856	−0.5764	1409.34
0.9252	0.879541	−0.4152	1410.43
1.0000	0.880501	0.0000	1411.28
T = 298.15 K			
0.0000	0.817494	0.000	1340.79
0.0443	0.825583	0.0611	1346.54
0.1134	0.835451	0.1455	1354.35
0.1775	0.842552	0.1995	1360.30
0.2062	0.845267	0.2133	1362.66
0.2457	0.848634	0.2199	1365.88
0.2868	0.851755	0.2114	1368.49
0.3202	0.85405	0.1924	1370.56
0.3721	0.857258	0.1415	1373.49
0.4104	0.859387	0.0880	1376.01
0.4628	0.862021	−0.0034	1378.32
0.5233	0.864719	−0.1291	1381.41
0.5674	0.866485	−0.2293	1383.24
0.6326	0.86882	−0.3765	1385.75
0.6872	0.870546	−0.4863	1387.41
0.7436	0.872124	−0.5695	1388.89
0.8251	0.874057	−0.5988	1390.84
0.8672	0.874899	−0.5530	1391.57
0.9252	0.875886	−0.3984	1392.63
1.0000	0.876862	0.0000	1393.41
T = 303.15 K			
0.0000	0.813986	0.0000	1322.14
0.0443	0.82209	0.0486	1328.05
0.1134	0.831952	0.1247	1336.02
0.1775	0.839032	0.1793	1342.06
0.2062	0.841736	0.1948	1344.47
0.2457	0.845087	0.2050	1347.48
0.2868	0.848194	0.1997	1350.35
0.3202	0.850479	0.1832	1352.43
0.3721	0.853673	0.1359	1355.51
0.4104	0.855795	0.0836	1357.94
0.4628	0.858422	−0.0076	1360.35
0.5233	0.861118	−0.1371	1363.83
0.5674	0.862885	−0.2420	1365.61
0.6326	0.865224	−0.3982	1367.88
0.6872	0.866953	−0.5156	1369.62
0.7436	0.868535	−0.6073	1371.06
0.8251	0.870468	−0.643	1372.89
0.8672	0.871306	−0.5959	1372.72
0.9252	0.872281	−0.4312	1374.76
1.0000	0.873227	0.0000	1375.60
T = 308.15 K			
0.0000	0.810476	0.0000	1303.63
0.0443	0.818533	0.0600	1306.61
0.1134	0.82836	0.1438	1317.86
0.1775	0.835433	0.1965	1323.99
0.2062	0.838138	0.2093	1326.39
0.2457	0.841493	0.2141	1329.48
0.2868	0.844603	0.2035	1332.40
0.3202	0.846892	0.1816	1334.60
0.3721	0.850091	0.1262	1337.79
0.4104	0.852214	0.0693	1339.95
0.4628	0.854842	−0.0281	1342.92
0.5233	0.857535	−0.1619	1345.86
0.5674	0.859297	−0.2674	1347.82
0.6326	0.861626	−0.4218	1350.18
0.6872	0.863346	−0.5361	1351.86

Table 4. Continued

	ρ_{12}	V^E_{12}	u_{12}
x_1	$g\cdot cm^{-3}$	$cm^3\cdot mol^{-1}$	$m\cdot s^{-1}$
0.7436	0.864918	−0.6228	1353.38
0.8251	0.866837	−0.6494	1355.29
0.8672	0.86767	−0.5987	1356.02
0.9252	0.86864	−0.4297	1357.00
1.0000	0.869588	0.0000	1357.91

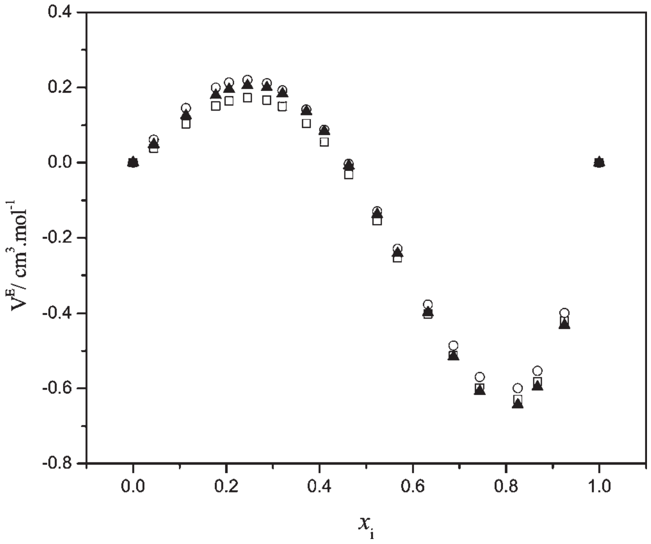

Figure 1. Excess molar volumes, V^E_{12} values of *J. curcas* biodiesel (1) + diesel fuel no. 2 (2) at temperatures of □, 288.15 K; ○, 298.15 K; ▲, 303.15 K and atmospheric pressure.

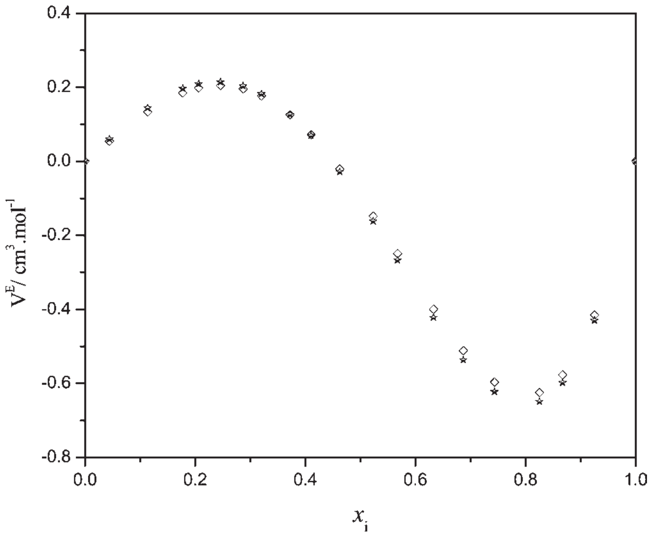

Figure 2. Excess molar volumes, V^E_{12} values of *J. curcas* biodiesel (1) + diesel fuel no. 2 (2) at temperatures of ◇, 293.15 K; ★, 308.15 K and atmospheric pressure.

where x_1, M_1, and ρ_1 are the mole fraction, molar mass, and density of the component 1, and ρ_{mix} is the density of the mixture, which are all recorded in Table 4 and plotted in Figures 1 and 2.

We are unaware of any existing V^E_{12} data for the studied binary mixtures with which to compare our results. The densities of the pure biodiesel and diesel as well as their mixture (blend) are decreasing linearly as a function of temperature, while the density of the mixture (blend) is increasing linearly with the increase in mole fraction of *J. curcas* biodiesel and vice versa. Though a substantial decrease in density has been observed with jatropha/diesel blend, still the densities are quite higher than those of diesel. The V^E_{12} values change sign from positive to negative with increasing mole fraction of biodiesel. Further, V^E_{12} values for the studied mixture are more negative at higher temperatures as compared to lower temperatures.

CONCLUSION

The *J. curcas* biodiesel was prepared, and the composition of this biodiesel was analyzed by gas chromatography with FID. Further, densities (ρ_{12}) and speeds of sound (u_{12}) of *J. curcas* biodiesel (1) + diesel fuel (2) binary mixture were measured as a function of composition at temperatures ranging from T = (288.15 to 308.15) K and atmospheric pressure. The observed data were utilized to evaluate the excess molar volumes, V^E_{12} . It has been observed that the densities (ρ) of the pure *J. curcas* biodiesel and diesel fuel no. 2 as well as their mixture (blend) decreased linearly as a function of temperature, while the density of the mixture (blend) increased linearly with increase in mole fraction of *J. curcas* biodiesel.

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