Journal of chemical & engineering data

Article

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Speed of Sound, Density, and Derivative Properties of Methyl Oleate and Methyl Linoleate under High Pressure

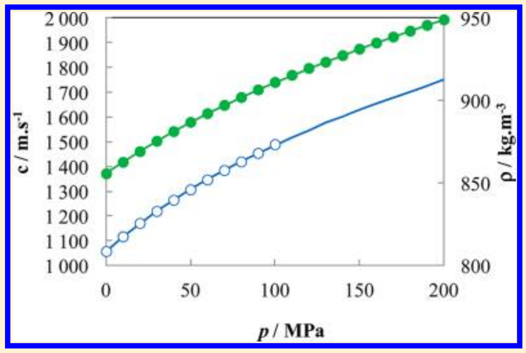
El Hadji I. Ndiaye,[†] Matthieu Habrioux,[†] João A. P. Coutinho,[‡] Márcio L. L. Paredes,[§] and Jean Luc Daridon*,[†]

[†]Laboratoire des Fluides Complexes et leurs Réservoirs, Faculté des Sciences et Techniques, UMR 5150, Université de Pau, BP 1155, 64013 Pau Cedex, France

[‡]CICECO, Chemistry Department, University of Aveiro, Campus de Santiago, 3810-193 Aveiro, Portugal

[§]Programa de Pós, Graduação em Engenharia Química, Universidade do Estado do Rio de Janeiro, Rua São Francisco Xavier, 524, Maracanã, Rio de Janeiro-RJ, Brasil

ABSTRACT: Speeds of sound were measured for methyl oleate and methyl linoleate ($C_{15}H_{30}O_2$) at pressures up to 200 MPa along isotherms ranging from (283.15 to 393.15) K. Additional density measurements were carried out by using a U-tube densimeter up to 100 MPa from (293 to 393) K. From the integration of speed of sound, density was evaluated up to 200 MPa, and the isentropic compressibility was determined in the same $p-T$ domain. A correlation that represents both the density and the speed of sound within their experimental uncertainties is reported to evaluate both the volume and its derivatives with respect to pressure (isothermal compressibility) and temperature (isobaric expansion).



1. INTRODUCTION

Biodiesels consist of alkyl esters of fatty acid mixtures produced by transesterification of various sources of biomass feedstock with a short chain alcohol such as methanol or ethanol. Rapeseed, sunflower, and soybean are the main biomass resources for biodiesels. However, a wide variety of regionally crops, such as palm, peanut, and coconut oils, can as well be used to produce biodiesels. Waste animal fats or other vegetable oils are also considered as raw material for biodiesel production. Most of fats and oils used for biodiesel generation contain straight chain aliphatic carboxylic acids with an even number of carbon atoms ranging between 10 and 24.[1] Each source has its own fatty acid composition. The chain may differ in length as well as in its degree of unsaturation. Consequently, the composition of fatty acid methyl esters may change a lot from one biodiesel to another according to the raw material sources. Fatty acid distribution affects thermophysical properties of biodiesels and influences engine efficiency as well as harmful gas emissions.[2] Among all of the thermophysical properties, density, isentropic compressibility, and viscosity, and cold flow properties (cloud point, cold filter plugging point, pour point) have a strong influence on engine efficiency as they directly affect the injection process. Cold flow properties can cause the fuel to gel[3,4] in the fuel delivery system during cold weather. Fuel viscosity acts on the pressure loss in the injector feed pipe[5] and therefore affects the quantity of fuel injected during each injection, while density is the main property that influences the conversion of volume flow rate into mass flow rate.[5] Finally, isentropic compressibility affects the wave velocity and consequently the fuel injection timing.[6]

To predict such thermophysical properties of biodiesels as a function of fatty acid profile, it is essential to have reliable experimental data concerning pure components. Therefore, we have initiated a program of measurement of several thermophysical properties of pure fatty acid methyl (or ethyl) esters in the framework of a collaborative program between the Universities of Aveiro (Portugal), Pau (France), and Rio de Janeiro (Brazil), and attempts will be made to correlate these data as a function of molecular structure.

The present work concerns the volumetric characterization of methyl oleate (MeC18:1) and methyl linoleate (MeC18:2) as a function of pressure from speed of sound measurements carried out up to 200 MPa over a substantial range of temperatures (283 to 393) K. It follows on other investigations of pure methyl caprate and ethyl caprate,[7] methyl myristate, methyl palmitate and ethyl myristate[8,9] and ethyl laurate[9] in the high pressure range. These methyl esters are produced from transesterification of oleic and linoleic acids that are predominant (70 to 90) % in the most common vegetable oils

Table 1. Sample Description

chemical name	shorthand designation	source	initial mole fraction purity	purification method
methyl oleate	MeC 18:1	Sigma-Aldrich	0.99	none
methyl linoleate	MeC 18:2	Sigma-Aldrich	0.99	none

Received: June 4, 2013
Accepted: July 10, 2013
Published: July 26, 2013

Table 2. Experimental Values of Speed of Sound c at Temperatures T and Pressures p for the Liquid Methyl Oleate and Methyl Linoleate[a]

p	T	c	T	c	T	c	T	c
MPa	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$
				Methyl Oleate				
0.1013	283.15	1443.2	303.15	1370.5	323.15	1301.9	343.15	1235.8
10	283.15	1486.1	303.15	1416.8	323.15	1352.1	343.15	1289.0
20	283.15	1526.5	303.15	1460.0	323.15	1397.7	343.15	1338.1
30	283.15	1564.5	303.15	1502.0	323.15	1441.1	343.15	1383.7
40	283.15	1600.5	303.15	1540.9	323.15	1482.3	343.15	1426.0
50	283.15	1634.5	303.15	1578.3	323.15	1521.2	343.15	1466.7
60	283.15	1668.6	303.15	1613.7	323.15	1558.5	343.15	1505.4
70	283.15	1701.0	303.15	1647.0	323.15	1593.5	343.15	1542.2
80	283.15	1732.7	303.15	1678.8	323.15	1627.2	343.15	1575.4
90	283.15	1762.7	303.15	1709.5	323.15	1659.3	343.15	1609.4
100	283.15	1790.8	303.15	1739.7	323.15	1690.2	343.15	1641.6
110	283.15	1819.3	303.15	1768.5	323.15	1719.3	343.15	1672.6
120	283.15	1846.0	303.15	1796.4	323.15	1747.7	343.15	1701.9
130	283.15	1872.5	303.15	1821.2	323.15	1775.4	343.15	1729.9
140	283.15	1897.8	303.15	1848.1	323.15	1801.6	343.15	1757.3
150	283.15	1923.3	303.15	1874.2	323.15	1827.1	343.15	1783.8
160	283.15	1947.8	303.15	1899.0	323.15	1853.0	343.15	1809.7
170	283.15	1971.2	303.15	1923.1	323.15	1878.4	343.15	1834.3
180	283.15	1994.3	303.15	1947.0	323.15	1902.6	343.15	1859.7
190	283.15	2016.8	303.15	1970.3	323.15	1925.8	343.15	1883.4
200	283.15	2036.4	303.15	1992.7	323.15	1949.2	343.15	1905.8
0.1013	363.15	1170.9	383.15	1108.9	393.15	1078.5		
10	363.15	1228.2	383.15	1170.4	393.15	1142.4		
20	363.15	1280.6	383.15	1226.5	393.15	1200.0		
30	363.15	1329.3	383.15	1277.8	393.15	1252.0		
40	363.15	1374.0	383.15	1324.6	393.15	1299.9		
50	363.15	1414.4	383.15	1367.4	393.15	1343.8		
60	363.15	1454.6	383.15	1407.7	393.15	1384.5		
70	363.15	1492.2	383.15	1446.1	393.15	1423.6		
80	363.15	1527.5	383.15	1481.7	393.15	1460.2		
90	363.15	1563.5	383.15	1516.9	393.15	1495.7		
100	363.15	1597.2	383.15	1550.2	393.15	1529.7		
110	363.15	1629.1	383.15	1583.4	393.15	1562.8		
120	363.15	1659.3	383.15	1615.8	393.15	1593.9		
130	363.15	1687.5	383.15	1645.9	393.15	1624.5		
140	363.15	1715.4	383.15	1674.2	393.15	1654.6		
150	363.15	1741.8	383.15	1701.5	393.15	1683.0		
160	363.15	1766.9	383.15	1729.4	393.15	1710.3		
170	363.15	1793.1	383.15	1755.3	393.15	1736.2		
180	363.15	1817.7	383.15	1781.4	393.15	1761.2		
190	363.15	1841.2	383.15	1805.7	393.15	1785.3		
200	363.15	1864.9	383.15	1828.3	393.15	1809.7		
				Methyl Linoleate				
0.1013	283.15	1455.0	303.15	1382.9	323.15	1312.6	343.15	1245.1
10	283.15	1498.2	303.15	1428.9	323.15	1361.4	343.15	1297.9
20	283.15	1539.0	303.15	1472.2	323.15	1407.2	343.15	1346.9
30	283.15	1577.6	303.15	1513.6	323.15	1450.8	343.15	1392.6
40	283.15	1614.3	303.15	1552.0	323.15	1491.3	343.15	1435.4
50	283.15	1649.2	303.15	1588.6	323.15	1530.3	343.15	1475.1
60	283.15	1682.6	303.15	1623.5	323.15	1567.2	343.15	1513.2
70	283.15	1714.0	303.15	1656.3	323.15	1601.4	343.15	1549.4
80	283.15	1745.2	303.15	1688.3	323.15	1634.3	343.15	1583.9
90	283.15	1774.4	303.15	1719.1	323.15	1666.2	343.15	1616.7
100	283.15	1803.2	303.15	1748.7	323.15	1697.2	343.15	1648.6
110	283.15	1830.9	303.15	1776.9	323.15	1726.4	343.15	1679.0
120	283.15	1857.8	303.15	1804.4	323.15	1754.6	343.15	1708.5
130	283.15	1884.6	303.15	1831.3	323.15	1782.4	343.15	1736.1

Table 2. continued

p	T	c	T	c	T	c	T	c
MPa	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$	K	$m \cdot s^{-1}$
				Methyl Linoleate				
140	283.15	1909.9	303.15	1857.0	323.15	1808.7	343.15	1763.4
150	283.15	1934.7	303.15	1883.6	323.15	1834.7	343.15	1789.7
170	283.15		303.15	1932.3	323.15	1884.5	343.15	1840.9
190	283.15		303.15	1979.7	323.15	1931.7	343.15	1889.3
210	283.15		303.15	2023.6	323.15	1977.6	343.15	1935.4
0.1013	363.15	1180.3	383.15	1118.2	393.15	1088.0		
10	363.15	1237.0	383.15	1178.7	393.15	1150.9		
20	363.15	1288.6	383.15	1233.8	393.15	1207.3		
30	363.15	1336.6	383.15	1283.1	393.15	1258.5		
40	363.15	1381.1	383.15	1329.0	393.15	1304.7		
50	363.15	1422.6	383.15	1372.3	393.15	1348.4		
60	363.15	1462.6	383.15	1413.2	393.15	1389.4		
70	363.15	1500.2	383.15	1452.3	393.15	1428.7		
80	363.15	1536.1	383.15	1489.5	393.15	1466.7		
90	363.15	1570.2	383.15	1525.2	393.15	1503.1		
100	363.15	1602.5	383.15	1558.2	393.15	1537.7		
110	363.15	1634.0	383.15	1590.3	393.15	1570.0		
120	363.15	1663.9	383.15	1621.1	393.15	1600.8		
130	363.15	1693.0	383.15	1650.8	393.15	1630.7		
140	363.15	1720.3	383.15	1679.1	393.15	1660.1		
150	363.15	1747.4	383.15	1707.1	393.15	1688.0		
170	363.15	1799.1	383.15	1759.8	393.15	1740.7		
190	363.15	1847.3	383.15	1808.6	393.15	1790.0		
210	363.15	1894.7	383.15	1855.8	393.15	1838.1		

[a]Standard uncertainties u are $u(T) = 0.1$ K, $u(p) = 0.01$ MPa up to 100 MPa, and $u(p) = 0.1$ MPa between (100 and 210) MPa and the combined expanded uncertainties U_c (level of confidence = 0.95) are $U_c(c) = 0.002c$ up to 100 MPa, $U_c(c) = 0.003c$ between (100 and 210) MPa.

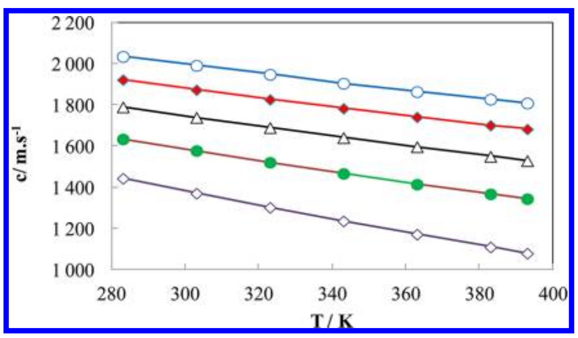


Figure 1. Speed of sound c in liquid methyl oleate as a function of temperature along various isobars. Purple ◇, 0.1 MPa; green ●, 50 MPa; black △, 100 MPa; red ◆, 150 MPa; blue ○, 200 MPa.

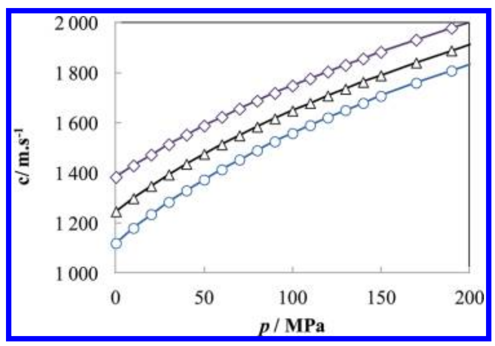


Figure 2. Speed of sound c in liquid methyl linoleate as a function of pressure along various isotherms. Purple ◇, 303.15 K; black △, 343.15 K; blue ○, 383.15 K.

(rapeseed, soybean, sunflower, etc.). They differ from their degree of unsaturation. The methyl oleate presents one double bond in the alkyl chain, whereas methyl linoleate has two.

2. EXPERIMENTAL SECTION

2.1. Materials. Table 1 shows the sample descriptions of methyl oleate (*cis*-9-octyldecenoic acid, methyl ester, CAS Number: 112-62-9, molar mass: 296.49 $g \cdot mol^{-1}$), methyl linoleate (*cis*-9,*cis*-12-octadecadienoic acid, methyl ester, CAS: 112-63-0, molar mass: 294.4721 $g \cdot mol^{-1}$) used in the present work.

2.2. Speed of Sound Measurement. The speed of sound was measured in an acoustic cell designed for working up to 210 MPa using a pulse-echo method. The apparatus, which has been described in detail previously,[7] consists of an acoustic probe made up of two 3 MHz piezo-ceramic discs. These electromechanical transducers were arranged facing each other at both ends of a hollow cylindrical support. One transducers acts as transmitter whereas the other works as a receiver. The speed of sound was evaluated from the measurement of the time delay between the transmitted pulse and the first echo by means of a numerical oscilloscope. The path length needed for calculating speed of sound was determined by calibration at different pressure and temperature with purified liquid water using the data of Wilson[10] and Baltasar et al.[11] The entire acoustic probe was immersed into the liquid sample within a high pressure vessel closed at one end by a plug having two electric connectors. The pressure was generated by a high pressure volumetric pump and transmitted to the cell by the studied liquid itself. It was measured by using two pressure gauges fixed on the circuit linking the pump to the measurement cell. One was calibrated in the full high-pressure scale (0.1 to 200) MPa with an uncertainty of 0.1 MPa, whereas

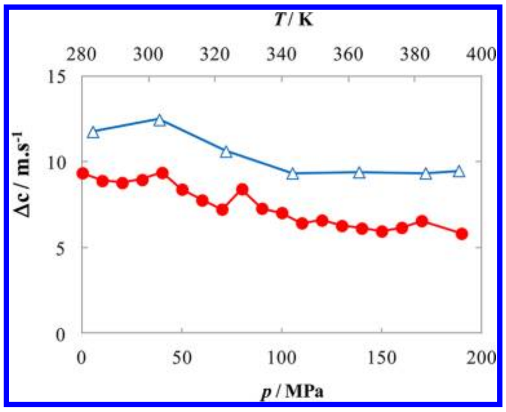


Figure 3. Deviations $\Delta c = c(\text{MeC18:2}) - c(\text{MeC18:1})$ between speed of sound data measured in methyl linoleate and methyl oleate. Red ●, isotherm 343.15 K; blue △, isobar 100 MPa.

Table 3. Parameters of eq 2 for Methyl Oleate and Methyl Linoleate

parameters	methyl oleate	methyl linoleate
A_0	$1.68680 \cdot 10^{-7}$	$1.49989 \cdot 10^{-7}$
A_1	$-1.85150 \cdot 10^{-10}$	$-7.77100 \cdot 10^{-11}$
A_2	$2.87672 \cdot 10^{-12}$	$2.61474 \cdot 10^{-12}$
A_3	$-3.09590 \cdot 10^{-15}$	$-3.25990 \cdot 10^{-15}$
B	$1.57821 \cdot 10^{-9}$	$1.17053 \cdot 10^{-9}$
C	$-4.51550 \cdot 10^{-12}$	$-2.73000 \cdot 10^{-12}$
D	$8.04761 \cdot 10^{-15}$	$4.06566 \cdot 10^{-15}$
E_1	$-1.50038 \cdot 10^{-3}$	$-1.56365 \cdot 10^{-3}$
F	$6.98325 \cdot 10^{-3}$	$6.04418 \cdot 10^{-3}$
	Deviations[a]	
AD%	$4.3 \cdot 10^{-4}$	$4.4 \cdot 10^{-3}$
AAD%	$5.1 \cdot 10^{-2}$	$4.2 \cdot 10^{-2}$
MD%	$1.4 \cdot 10^{-1}$	$1.5 \cdot 10^{-1}$

[a]AD = average deviation; AAD = absolute average deviation; MD = maximum deviation.

the other was only calibrated between (0.1 and 100) MPa to reduce the uncertainty to 0.01 MPa in this pressure range. The temperature of the acoustic cell was thermostatically controlled by entirely immersing it in a thermoregulated bath. The temperature of the liquid inside the cell was measured with an uncertainty of 0.1 K by a Pt100 probe in contact with the liquid sample. The expanded uncertainty for speed of measurements was estimated to be 0.2 % between (0.1 and 100) MPa and 0.3 % between (100 and 200) MPa by using the law of propagation of standard uncertainty[12] and by considering the conventional coverage factor $k_P = 2$ ($P = 95\%$).

2.3. Density Measurement. Density was measured up to 100 MPa by using a U-shape tube densimeter related to a high-pressure volumetric pump. The apparatus was calibrated with vacuum and a reference liquid. According to the method proposed by Comuñas et al.,[13] purified liquid water[14] was used as reference liquid over the entire temperature and pressure range apart from temperatures near or above the boiling point of water ($p = 0.1013$ MPa, $T \geq 363$ K). In this particular range, decane (mole fraction purity ≥ 0.99) was selected as reference liquid instead of water as its density is well-known[15] at atmospheric pressure over wide temperature intervals. The temperature was measured thanks to a HBM pressure gauge (0.1 % of uncertainty) located between the measurement cell and the volumetric pump. Taking into account the uncertainty in measurements of oscillation period, temperature, and pressure as well the error in the calibration method, the overall experimental uncertainty in the reported density values is estimated by Comuñas et al.[13] to be ± 0.5 $\text{kg} \cdot \text{m}^{-3}$ (0.06 %) in the pressure range investigated.

To extend the density data to higher pressures than 100 MPa, the change in density ρ with respect to pressure was evaluated from the integration of sound speed measurement up to 210 MPa:[15,16]

$$\rho(p, T) - \rho(p_{\text{ref}}, T) = \int_{p_{\text{ref}}}^{p} \frac{1}{c^2} \text{d}p + T \int_{p_{\text{ref}}}^{p} \frac{\alpha_P^2}{c_P} \text{d}p \quad (1)$$

where α_P is the isobaric expansion coefficient and c_p the heat capacity at constant pressure. P_{ref} is a reference pressure where the density and heat capacity are known. In this work it corresponds to the atmospheric pressure. The first integral of eq 1 represents the main contribution to the change of density with pressure. It is evaluated directly from the speed of sound measurements by first correlating $1/c^2$ with temperature and

Table 4. Deviations between Literature Data and Value Interpolated by eqs 1 and 4 to 7[a]

			methyl oleate			methyl linoleate		
ref	T range/K	P range/MPa	AD%	AAD%	MD%	AD%	AAD%	MD%
			Speed of Sound					
19	293−313	0.1013	−0.18	0.18	0.22	0.12	0.12	0.15
20	278−338	0.1013	−0.04	0.05	0.08	0.09	0.09	0.12
21	283−373	0.1013	−0.09	0.12	0.21	0.06	0.08	0.16
22	288−343	0.1013	−0.31	0.31	0.34			
			Density					
19	293−313	0.1013	−0.03	0.03	0.03	−0.15	0.15	0.17
23	298	0.1013	−0.4			−0.47		
24	298	0.1013	−0.02			−0.10		
25	298	0.1013	−0.04			−0.11		
20	278−338	0.1013	−0.01	0.02	0.04	−0.04	0.04	0.07
26	293−363	0.1013	−0.02	0.02	0.06	−0.09	0.09	0.13
27	293−363	0.1013	0.005	0.008	0.05			
28	290−390	0.08−50	−0.19	0.19	0.26	−0.13	0.13	0.31
29-1	297−367	0.5−60				−0.06	0.06	0.09
29-2	297−337	0.4−130				−0.09	0.09	0.15

[a]AD = average deviation; AAD = absolute average deviation; MD = maximum deviation.

Table 5. Values of Densities ρ at Temperatures T and Pressures p Measured in Liquid Methyl Oleate and Methyl Linoleate by Using a U-Tube Densimeter[a]

p/MPa	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}
						Methyl Oleate						
0.1013	293.15	873.8	303.15	866.3	313.15	859.6	323.15	852.4	333.15	844.8	343.15	837.8
10	293.15	880.0	303.15	872.8	313.15	865.8	323.15	858.9	333.15	851.7	343.15	845.2
20	293.15	885.3	303.15	878.4	313.15	871.7	323.15	865.1	333.15	858.3	343.15	851.9
30	293.15	890.6	303.15	883.9	313.15	877.5	323.15	870.9	333.15	864.2	343.15	858.1
40	293.15	895.5	303.15	888.9	313.15	882.5	323.15	876.1	333.15	869.7	343.15	864.0
50	293.15	900.1	303.15	893.7	313.15	887.5	323.15	881.1	333.15	875.0	343.15	869.5
60	293.15	904.5	303.15	898.3	313.15	892.2	323.15	886.2	333.15	880.1	343.15	874.6
70	293.15	908.9	303.15	902.7	313.15	896.7	323.15	891.0	333.15	884.9	343.15	879.5
80	293.15	912.6	303.15	906.9	313.15	901.2	323.15	895.5	333.15	889.4	343.15	884.4
90	293.15	916.9	303.15	910.9	313.15	905.2	323.15	899.7	333.15	893.9	343.15	889.0
100	293.15	920.7	303.15	915.4	313.15	909.7	323.15	904.1	333.15	898.2	343.15	893.4
0.1013	353.15	830.4	363.15	823.2	373.15	815.8	383.15	808.4	393.15	800.8		
10	353.15	838.0	363.15	831.3	373.15	824.2	383.15	817.4	393.15	810.2		
20	353.15	845.1	363.15	838.7	373.15	832.0	383.15	825.5	393.15	818.5		
30	353.15	851.5	363.15	845.5	373.15	839.1	383.15	832.7	393.15	826.2		
40	353.15	857.5	363.15	851.8	373.15	845.7	383.15	839.5	393.15	833.2		
50	353.15	863.3	363.15	857.7	373.15	851.8	383.15	845.9	393.15	839.6		
60	353.15	868.7	363.15	863.1	373.15	857.5	383.15	851.8	393.15	845.7		
70	353.15	873.8	363.15	868.4	373.15	862.9	383.15	857.5	393.15	851.7		
80	353.15	878.7	363.15	873.3	373.15	868.2	383.15	862.8	393.15	857.2		
90	353.15	883.6	363.15	878.1	373.15	873.0	383.15	867.9	393.15	862.4		
100	353.15	888.2	363.15	882.9	373.15	878.1	383.15	873.2	393.15	867.6		
						Methyl Linoleate						
0.1013	293.15	885.2	303.15	877.7	313.15	870.9	323.15	863.7	333.15	856.3	343.15	849.1
10	293.15	891.2	303.15	884.0	313.15	877.1	323.15	870.1	333.15	863.0	343.15	856.2
20	293.15	896.6	303.15	889.6	313.15	882.9	323.15	876.3	333.15	869.5	343.15	863.0
30	293.15	901.8	303.15	895.0	313.15	888.5	323.15	882.0	333.15	875.4	343.15	869.3
40	293.15	906.8	303.15	900.1	313.15	893.7	323.15	887.3	333.15	881.3	343.15	875.1
50	293.15	911.4	303.15	905.0	313.15	898.8	323.15	892.4	333.15	886.5	343.15	880.8
60	293.15	915.9	303.15	909.5	313.15	903.3	323.15	897.4	333.15	891.5	343.15	885.9
70	293.15	920.2	303.15	914.0	313.15	908.0	323.15	902.1	333.15	896.4	343.15	890.9
80	293.15	924.0	303.15	918.1	313.15	912.2	323.15	906.7	333.15	900.9	343.15	895.7
90	293.15	928.2	303.15	922.2	313.15	916.6	323.15	910.9	333.15	905.4	343.15	900.3
100	293.15	931.3	303.15	926.2	313.15	920.6	323.15	915.1	333.15	910.0	343.15	904.7
0.1013	353.15	841.8	363.15	834.7	373.15	826.5	383.15	819.3	393.15	811.9		
10	353.15	849.3	363.15	842.2	373.15	835.3	383.15	828.2	393.15	820.9		
20	353.15	856.2	363.15	849.5	373.15	842.9	383.15	836.2	393.15	829.3		
30	353.15	862.8	363.15	856.4	373.15	849.9	383.15	843.5	393.15	837.0		
40	353.15	868.9	363.15	862.6	373.15	856.4	383.15	850.2	393.15	844.1		
50	353.15	874.6	363.15	868.4	373.15	862.4	383.15	856.8	393.15	850.5		
60	353.15	879.9	363.15	873.9	373.15	868.3	383.15	862.7	393.15	856.7		
70	353.15	885.2	363.15	879.1	373.15	873.9	383.15	868.2	393.15	862.4		
80	353.15	890.0	363.15	884.1	373.15	879.3	383.15	873.5	393.15	867.9		
90	353.15	894.7	363.15	889.0	373.15	884.3	383.15	878.7	393.15	873.0		
100	353.15	899.3	363.15	893.9	373.15	889.1	383.15	883.9	393.15	878.1		

[a]Standard uncertainties u are $u(T) = 0.1$ K and $u(p) = 0.01$ MPa, and the combined expanded uncertainty U_c (level of confidence = 0.95) is $U_c(\rho) = 0.5$ kg·m^{-3}.

pressure and then by integrating it analytically. For that purpose, a rational function with nine adjustable parameters and with a denominator limited to the first degree in both temperature and pressure was considered:

$$\frac{1}{c^2} = \frac{A_0 + A_1T + A_2T^2 + A_3T^3 + Bp + Cp^2 + Dp^3}{1 + ET + Fp} \qquad (2)$$

The second integral, the value of which represents only a few percent of the first, is evaluated iteratively using a predictor-corrector procedure presented in details in previous papers.[16,17] The heat capacity data needed to initiate the procedure were taken at atmospheric pressure[18] and were correlated with temperature in the range investigated:

$$c_p(\mathrm{MeC18{:}1})/\mathrm{J{\cdot}K^{-1}{\cdot}kg^{-1}} = 1.128\cdot 10^3 + 2.661T + 8.864\cdot 10^{-4}T^2 \qquad (3)$$

$$c_p(\mathrm{MeC18{:}2})/\mathrm{J{\cdot}K^{-1}{\cdot}kg^{-1}} = 8.868\cdot 10^2 + 3.367T \qquad (4)$$

dx.doi.org/10.1021/je4005323 | J. Chem. Eng. Data 2013, 58, 2345−2354

Table 6. Values of Densities ρ at Temperatures T and Pressures p Determined from the Integration of Speed of Sound Measurements in Liquid Methyl Oleate and Methyl Linoleate[a]

p/MPa	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}
						Methyl Oleate						
0.1013	303.15	866.6	323.15	852.2	343.15	837.8	363.15	823.2	383.15	808.3	393.15	800.8
10	303.15	872.6	323.15	858.9	343.15	845.1	363.15	831.2	383.15	817.3	393.15	810.2
20	303.15	878.3	323.15	865.1	343.15	851.9	363.15	838.7	383.15	825.4	393.15	818.7
30	303.15	883.7	323.15	870.9	343.15	858.2	363.15	845.5	383.15	832.8	393.15	826.5
40	303.15	888.8	323.15	876.4	343.15	864.1	363.15	851.9	383.15	839.7	393.15	833.6
50	303.15	893.6	323.15	881.6	343.15	869.7	363.15	857.9	383.15	846.1	393.15	840.2
60	303.15	898.2	323.15	886.5	343.15	874.9	363.15	863.5	383.15	852.1	393.15	846.4
70	303.15	902.6	323.15	891.2	343.15	879.9	363.15	868.8	383.15	857.7	393.15	852.2
80	303.15	906.8	323.15	895.7	343.15	884.7	363.15	873.8	383.15	863.1	393.15	857.7
90	303.15	910.9	323.15	900.0	343.15	889.2	363.15	878.6	383.15	868.1	393.15	862.9
100	303.15	914.8	323.15	904.1	343.15	893.6	363.15	883.2	383.15	873.0	393.15	867.9
110	303.15	918.6	323.15	908.1	343.15	897.8	363.15	887.6	383.15	877.6	393.15	872.6
120	303.15	922.2	323.15	911.9	343.15	901.8	363.15	891.9	383.15	882.1	393.15	877.2
130	303.15	925.7	323.15	915.6	343.15	905.7	363.15	895.9	383.15	886.3	393.15	881.6
140	303.15	929.2	323.15	919.2	343.15	909.5	363.15	899.9	383.15	890.4	393.15	885.8
150	303.15	932.5	323.15	922.7	343.15	913.1	363.15	903.7	383.15	894.4	393.15	889.8
160	303.15	935.7	323.15	926.1	343.15	916.6	363.15	907.4	383.15	898.3	393.15	893.8
170	303.15	938.9	323.15	929.3	343.15	920.0	363.15	910.9	383.15	902.0	393.15	897.6
180	303.15	941.9	323.15	932.5	343.15	923.4	363.15	914.4	383.15	905.6	393.15	901.2
190	303.15	944.9	323.15	935.6	343.15	926.6	363.15	917.8	383.15	909.1	393.15	904.8
200	303.15	947.8	323.15	938.7	343.15	929.8	363.15	921.0	383.15	912.5	393.15	908.3
						Methyl Linoleate						
0.1013	303.15	878.0	323.15	863.6	343.15	849.1	363.15	834.3	383.15	819.3	393.15	811.8
10	303.15	884.0	323.15	870.2	343.15	856.4	363.15	842.4	383.15	828.2	393.15	821.1
20	303.15	889.6	323.15	876.4	343.15	863.1	363.15	849.8	383.15	836.3	393.15	829.6
30	303.15	894.9	323.15	882.2	343.15	869.4	363.15	856.6	383.15	843.7	393.15	837.3
40	303.15	900.0	323.15	887.7	343.15	875.3	363.15	862.9	383.15	850.6	393.15	844.4
50	303.15	904.8	323.15	892.8	343.15	880.8	363.15	868.9	383.15	857.0	393.15	851.0
60	303.15	909.3	323.15	897.7	343.15	886.1	363.15	874.5	383.15	862.9	393.15	857.2
70	303.15	913.7	323.15	902.4	343.15	891.1	363.15	879.8	383.15	868.6	393.15	863.0
80	303.15	917.9	323.15	906.8	343.15	895.8	363.15	884.8	383.15	873.9	393.15	868.5
90	303.15	921.9	323.15	911.1	343.15	900.3	363.15	889.6	383.15	879.0	393.15	873.7
100	303.15	925.7	323.15	915.2	343.15	904.7	363.15	894.2	383.15	883.9	393.15	878.7
110	303.15	929.5	323.15	919.1	343.15	908.8	363.15	898.6	383.15	888.5	393.15	883.5
120	303.15	933.1	323.15	922.9	343.15	912.9	363.15	902.9	383.15	892.9	393.15	888.0
130	303.15	936.6	323.15	926.6	343.15	916.7	363.15	906.9	383.15	897.2	393.15	892.4
140	303.15	940.0	323.15	930.2	343.15	920.5	363.15	910.9	383.15	901.3	393.15	896.6
150	303.15	943.3	323.15	933.7	343.15	924.1	363.15	914.7	383.15	905.3	393.15	900.7
160	303.15	946.5	323.15	937.0	343.15	927.6	363.15	918.4	383.15	909.2	393.15	904.6
170	303.15	949.6	323.15	940.3	343.15	931.1	363.15	921.9	383.15	912.9	393.15	908.4
180	303.15	952.6	323.15	943.4	343.15	934.4	363.15	925.4	383.15	916.5	393.15	912.1
190	303.15	955.6	323.15	946.5	343.15	937.6	363.15	928.8	383.15	920.0	393.15	915.7
200	303.15	958.4	323.15	949.6	343.15	940.8	363.15	932.0	383.15	923.4	393.15	919.1

[a]Standard uncertainties u are $u(T)$ = 0.1 K, $u(p)$ = 0.01 MPa up to 100 MPa, and $u(p)$ = 0.1 MPa between (100 and 210) MPa, and the combined expanded uncertainties U_c (level of confidence = 0.95) are $U_c(\rho) = 0.001\rho$ up to 100 MPa and $U_c(\rho) = 0.002\rho$ between (100 and 210) MPa.

3. RESULTS AND DISCUSSION

Speed of sound measurements were undertaken along isotherms spaced out 20 K from (283.15 to 393.15) K at fixed conditions of pressure from atmospheric pressure to 200 MPa. The results are listed in Table 2 and are plotted as a function of temperature and pressure in Figures 1 and 2. Both components studied have very similar acoustic behaviors. The difference between sound speeds at 283.15 K at atmospheric pressure is only 12 m·s^{-1}, and this difference decreases with both increasing pressure and temperature as can be observed in Figure 3. The full sets of data were correlated using eq 1. The parameters obtained by a least-squares method are listed in Table 3 along with the average deviation (AD%), the average absolute deviation (AAD%), and the maximum deviation (MD%) for both components. As can be seen, the maximum deviation observed between the experimental speed-of-sound data and the correlation function is smaller than the experimental error. Moreover, an analysis of the average deviation shows that the correlation function does not introduce any systematic error. The quality of the measurements was checked by comparison with literature data. A survey of the literature reveals four sources[19−22] but limited to atmospheric pressure. A comparison between these

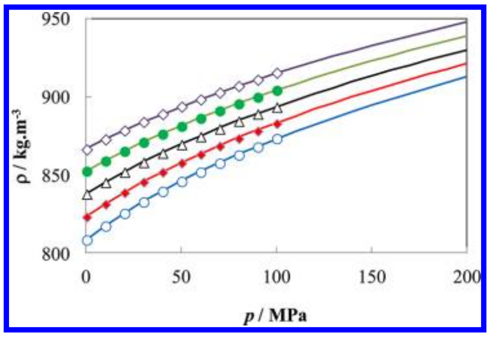


Figure 4. Comparison between densities of methyl oleate evaluated from speed of sound integration (line) and measured from a U-tube densimeter. Purple ◇, 303.15 K; green ●, 323.15 K; black △, 343.15 K; red ◆, 363.15 K; blue ○, 383.15 K.

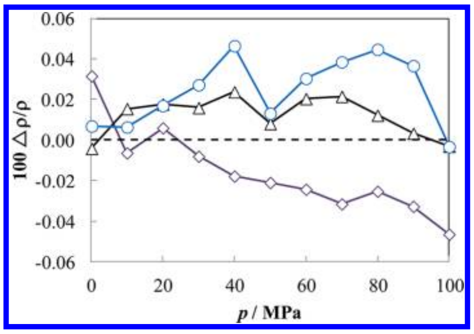


Figure 5. Relative deviations $\Delta\rho/\rho = \{\rho(\text{exp U-tube}) - \rho(\text{exp c})\}/\rho(\text{exp U-tube})$ between density data measured by a U-tube densimeter and determined from speed of sound measurements in liquid methyl linoleate. Purple ◇, 303.15 K; black △, 343.15 K; blue ○, 383.15 K.

Table 7. Parameters of eqs 5 to 8 and Deviations from Sound Speed and Density Data

parameters	methyl oleate	methyl linoleate
v_0	$8.64437\cdot10^{-4}$	$9.12887\cdot10^{-4}$
v_1	$1.21775\cdot10^{-6}$	$7.11802\cdot10^{-7}$
v_2	$-1.72500\cdot10^{-9}$	$-3.61610\cdot10^{-10}$
v_3	$2.83273\cdot10^{-12}$	$1.56379\cdot10^{-12}$
a_0	$1.13713\cdot10^{-5}$	$7.23693\cdot10^{-6}$
a_1	$5.95289\cdot10^{-7}$	$5.31998\cdot10^{-7}$
a_2	$-1.69530\cdot10^{-9}$	$-1.29370\cdot10^{-9}$
a_3	$1.94945\cdot10^{-12}$	$1.38553\cdot10^{-12}$
b_0	$3.92963\cdot10^{2}$	$3.73085\cdot10^{2}$
b_1	-1.31188	-1.18900
b_2	$1.21428\cdot10^{-3}$	$1.03984\cdot10^{-3}$
d	$4.28377\cdot10^{-8}$	$3.89658\cdot10^{-8}$
	Deviations[a]	
AD% for ρ	$1.2\cdot10^{-3}$	$5.9\cdot10^{-4}$
AAD% for ρ	$2.2\cdot10^{-3}$	$2.2\cdot10^{-3}$
MD% for ρ	$7.7\cdot10^{-3}$	$6.6\cdot10^{-3}$
AD% for c	$-1.1\cdot10^{-2}$	$-7.0\cdot10^{-3}$
AAD% for c	$9.6\cdot10^{-2}$	$9.1\cdot10^{-2}$
MD% for c	$2.0\cdot10^{-1}$	$3.0\cdot10^{-1}$

[a] AD = average deviation; AAD = absolute average deviation; MD = maximum deviation.

data and the value interpolated by eq 1 were listed in Table 4 for both components. The deviations observed in this table show an agreement between the different data sets within their experimental uncertainties.

Measurements of density were carried out with a U-tube densimeter up to 100 MPa along isotherms ranging from

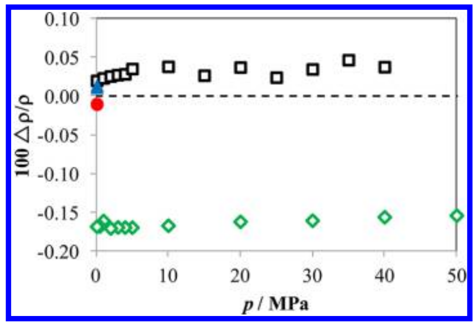


Figure 6. Relative deviations $\Delta\rho/\rho = \{\rho(\text{correlation}) - \rho(\text{literature})\}/\rho(\text{literature})$ between the proposed correlation (eqs 5 to 8) and literature data of methyl oleate at temperature ranging from (330 to 340) K. Blue ▲, Ott et al.,[20] 338.15 K; red ●, Pratas et al.,[26] 333.15 K; black □, Pratas et al.,[27] 333.15 K; green ◇, Outcalt,[28] 330 K.

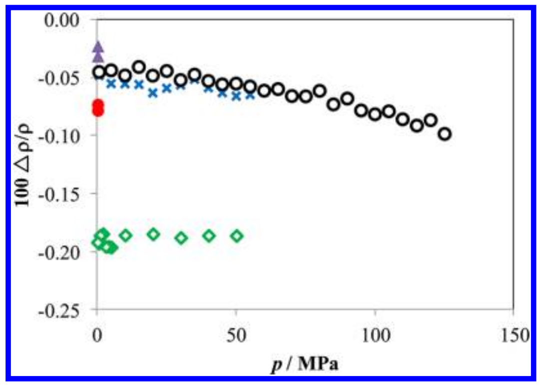


Figure 7. Relative deviations $\Delta\rho/\rho = \{\rho(\text{correlation}) - \rho(\text{literature})\}/\rho(\text{literature})$ between the proposed correlation (eqs 5 to 8) and literature data of methyl linoleate at temperature ranging from (330 to 340) K. Purple ▲, Ott et al.,[20] 338.15 K; red ●, Pratas et al.,[26] 333.15 K; green ◇, Outcalt,[28] 330 K; blue ×, Schedemann et al.,[29] 337.38 K; black ○, Schedemann et al.,[29] 337.38 K.

(293.15 to 393.15) K at 10 K intervals to get a more refined representation of the volumetric behavior as a function of temperature. High-pressure data were completed by the speed of sound integration in the same temperature range. The data obtained by both methods are reported in Tables 5 and 6 and plotted for methyl oleate in Figure 4. The estimation of the density trend with pressure evaluated from speed of integration matches perfectly the U-tube measurements. In the common range of pressure covered, a comparison of density data shows a satisfactory agreement with a maximum deviation that does not exceed 0.08 % for methyl oleate and 0.09 % for methyl linoleate. Moreover, by comparing the average (−0.016 % for MeC 18:1 and 0.010 % for MeC 18:2) and absolute average deviations (0.020 % for MeC 18:1 and 0.028 % for MeC 18:2), no systematic error appears between the two methods. Finally, it can be observed in Figure 5 that deviation between the two sets of data does not systematically increase with pressure.

This good agreement reflects the overall consistency between the speed of sound and the density measurements and confirms the ability to measure density under high pressure by the acoustic technique.

The volumetric behavior of both components studied was represented by the following equation:

$$v = v_{\text{ref}} - d(p - p_{\text{ref}}) + (bd - a)\ln\left(\frac{p + b}{p_{\text{ref}} + b}\right) \qquad (5)$$

dx.doi.org/10.1021/je4005323 | J. Chem. Eng. Data 2013, 58, 2345−2354

Table 8. Values of Isentropic Compressibility κ_S in Liquid Methyl Oleate and Methyl Linoleate at Temperatures T and Pressures p[a]

p / MPa	T / K	κ_S / GPa^{-1}	T / K	κ_S / GPa^{-1}	T / K	κ_S / GPa^{-1}	T / K	κ_S / GPa^{-1}	T / K	κ_S / GPa^{-1}	T / K	κ_S / GPa^{-1}
						Methyl Oleate						
0.1013	303.15	0.614	323.15	0.692	343.15	0.782	363.15	0.886	383.15	1.006	393.15	1.074
10	303.15	0.571	323.15	0.637	343.15	0.712	363.15	0.798	383.15	0.893	393.15	0.946
20	303.15	0.534	323.15	0.592	343.15	0.656	363.15	0.727	383.15	0.805	393.15	0.848
30	303.15	0.502	323.15	0.553	343.15	0.609	363.15	0.669	383.15	0.735	393.15	0.772
40	303.15	0.474	323.15	0.519	343.15	0.569	363.15	0.622	383.15	0.679	393.15	0.710
50	303.15	0.449	323.15	0.490	343.15	0.535	363.15	0.583	383.15	0.632	393.15	0.659
60	303.15	0.428	323.15	0.464	343.15	0.504	363.15	0.547	383.15	0.592	393.15	0.616
70	303.15	0.408	323.15	0.442	343.15	0.478	363.15	0.517	383.15	0.558	393.15	0.579
80	303.15	0.391	323.15	0.422	343.15	0.455	363.15	0.490	383.15	0.528	393.15	0.547
90	303.15	0.376	323.15	0.404	343.15	0.434	363.15	0.466	383.15	0.501	393.15	0.518
100	303.15	0.361	323.15	0.387	343.15	0.415	363.15	0.444	383.15	0.477	393.15	0.492
110	303.15	0.348	323.15	0.373	343.15	0.398	363.15	0.425	383.15	0.454	393.15	0.469
120	303.15	0.336	323.15	0.359	343.15	0.383	363.15	0.407	383.15	0.434	393.15	0.449
130	303.15	0.326	323.15	0.346	343.15	0.369	363.15	0.392	383.15	0.416	393.15	0.430
140	303.15	0.315	323.15	0.335	343.15	0.356	363.15	0.378	383.15	0.401	393.15	0.412
150	303.15	0.305	323.15	0.325	343.15	0.344	363.15	0.365	383.15	0.386	393.15	0.397
160	303.15	0.296	323.15	0.314	343.15	0.333	363.15	0.353	383.15	0.372	393.15	0.382
170	303.15	0.288	323.15	0.305	343.15	0.323	363.15	0.341	383.15	0.360	393.15	0.370
180	303.15	0.280	323.15	0.296	343.15	0.313	363.15	0.331	383.15	0.348	393.15	0.358
190	303.15	0.273	323.15	0.288	343.15	0.304	363.15	0.321	383.15	0.337	393.15	0.347
200	303.15	0.266	323.15	0.280	343.15	0.296	363.15	0.312	383.15	0.328	393.15	0.336
						Methyl Linoleate						
0.1013	303.15	0.596	323.15	0.672	343.15	0.760	363.15	0.860	383.15	0.976	393.15	1.041
10	303.15	0.554	323.15	0.620	343.15	0.693	363.15	0.776	383.15	0.869	393.15	0.919
20	303.15	0.519	323.15	0.576	343.15	0.639	363.15	0.709	383.15	0.786	393.15	0.827
30	303.15	0.488	323.15	0.539	343.15	0.593	363.15	0.653	383.15	0.720	393.15	0.754
40	303.15	0.461	323.15	0.507	343.15	0.554	363.15	0.608	383.15	0.666	393.15	0.696
50	303.15	0.438	323.15	0.478	343.15	0.522	363.15	0.569	383.15	0.620	393.15	0.646
60	303.15	0.417	323.15	0.454	343.15	0.493	363.15	0.535	383.15	0.580	393.15	0.604
70	303.15	0.399	323.15	0.432	343.15	0.467	363.15	0.505	383.15	0.546	393.15	0.568
80	303.15	0.382	323.15	0.413	343.15	0.445	363.15	0.479	383.15	0.516	393.15	0.535
90	303.15	0.367	323.15	0.395	343.15	0.425	363.15	0.456	383.15	0.489	393.15	0.507
100	303.15	0.353	323.15	0.379	343.15	0.407	363.15	0.435	383.15	0.466	393.15	0.481
110	303.15	0.341	323.15	0.365	343.15	0.390	363.15	0.417	383.15	0.445	393.15	0.459
120	303.15	0.329	323.15	0.352	343.15	0.375	363.15	0.400	383.15	0.426	393.15	0.439
130	303.15	0.318	323.15	0.340	343.15	0.362	363.15	0.385	383.15	0.409	393.15	0.421
140	303.15	0.308	323.15	0.329	343.15	0.349	363.15	0.371	383.15	0.394	393.15	0.405
150	303.15	0.299	323.15	0.318	343.15	0.338	363.15	0.358	383.15	0.379	393.15	0.390
170	303.15	0.282	323.15	0.299	343.15	0.317	363.15	0.335	383.15	0.354	393.15	0.363
190	303.15	0.267	323.15	0.283	343.15	0.299	363.15	0.316	383.15	0.332	393.15	0.341

[a]Standard uncertainties u are $u(T) = 0.1$ K, $u(p) = 0.01$ MPa up to 100 MPa, and $u(p) = 0.1$ MPa between (100 and 210) MPa, and the combined expanded uncertainties U_c (level of confidence = 0.95) are $U_c(\kappa_S) = 0.005\ \kappa_S$ up to 100 MPa and $U_c(\kappa_S) = 0.009\ \kappa_S$ between (100 and 200) MPa.

with:

$$v_{ref} = v_0 + v_1 T + v_2 T^2 + v_3 T^3 \tag{6}$$

$$a = a_0 + a_1 T + a_2 T^2 + a_3 T^3 \tag{7}$$

$$b = b_0 + b_1 T + b_2 T^2 \tag{8}$$

This equation expresses the change in volume with respect to pressure in a rational form:

$$\left(\frac{\partial v}{\partial p}\right)_T = -\frac{a + dp}{b + p} \tag{9}$$

It enables to calculate both density and sound speed data within their experimental uncertainties as heat capacities are known at the reference pressure (eqs 3 and 4). Parameters v_0, v_1, v_2, and v_3 were first adjusted to reflect atmospheric density. Coefficients a_i, b_i, and d were determined by minimizing the following objective function that intercorrelates density data and speed of sound measurements:

$$\text{Obj} = \sum_i^{N_{exp}} \left(\left(\frac{v_i^{exp}}{c_i^{exp}} \right)^2 + \left(\frac{\partial v}{\partial p} \right)_T^{cal} + \frac{T}{c_p^{cal}} \left(\frac{\partial v}{\partial T} \right)_p^{2,cal} \right)^2 \tag{10}$$

Table 7 lists the coefficients obtained by this procedure along with the AD, the AAD, and the MD with speed of sound and density data.

Observation of the deviations reveals an excellent agreement between the calculated and experimental densities with a maximum deviation that does not exceed 0.01 % for both compounds investigated. Moreover, the maximum deviation observed between calculated and experimental speed of sound is 0.3 %, which corresponds to the experimental error in the higher pressure range (100 to 200 MPa). This result demonstrates the capacity of eq 5 to predict both volume and its derivatives in an extended pressure range. Therefore it can be used to derive the isothermal compressibility as well as the isobaric expansion from a combination of the two sets of experimental measurements.

Density measurements were carried out previously in methyl oleate and methyl linoleate at atmospheric pressure by several authors.[19,20,23−26] A comparison between the proposed correlation (eq 6) and the data report by these authors shows a good agreement (Table 4). Only the data points reported by Komoda et al.[23] at 298 K for both components deviate significantly from the proposed correlation. At the opposite the correlation matches perfectly with the data of Ott et al.[20] with a maximum deviation of 0.04 % for methyl oleate and 0.07 % for methyl linoleate.

Density measurements performed in methyl oleate by using a vibrating U-tube densimeter were reported by Pratas et al.[27] and Outcalt[28] in a moderate pressure range (0.1 to 50 MPa). The comparison (Table 4 and Figure 6) shows significant systematic negative deviations between our correlation and data reported by Outcalt,[28] whereas a small positive deviation is observed in average with data of Pratas et al.[27] meaning that our data are located between those of literature data but closer than those reported by Pratas et al.[27] As for methyl linoleate, high-pressure measurements were reported by Outcalt.[28] Schedemann et al.[29] also provide two set of experimental data measured by a vibrating U-tube densimeter. The proposed correlation is slightly smaller than the measurements reported by Schedemann et al.[29] as can be observed in Table 4. The deviation do not exceed 0.1 % with the first set limited to 60 MPa and 0.15 % with the second that is extended to 130 MPa. As with methyl oleate, a systematic negative deviation is observed (Figure 7) with density data of methyl oleate reported by Outcalt.[28]

Finally, to complete the high-pressure characterization, isentropic compressibility was evaluated from the simultaneous knowledge of density and speed of sound:

$$\kappa_S = \frac{1}{\rho c^2} \tag{11}$$

The values determined in this way from density data obtained from speed of sound measurements are listed in Table 8.

4. CONCLUSIONS

Speeds of sound of liquid methyl oleate and methyl linoleate were measured over an extended pressure range between (0.1 and 200) MPa with temperature ranging from (283 to 393) K. Densities were also measured for the same compounds by using a U-tube densimeter up to 100 MPa and were determined between atmospheric pressure and 200 MPa by integration of speed of sound data. A satisfactory agreement was observed between both sets of density data. The measurements were used to evaluate the isentropic compressibility. Finally a correlation was proposed to represent both density and speed of sound data within their experimental uncertainty. The good agreement observed between the calculated and experimental data for both properties in the wide pressure range investigated confirms the consistency between the two set of thermophysical properties and proves its capacity to represent both the density and its derivative with respect to pressure (isothermal compressibility) and temperature (isobaric expansion).

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: jean-luc.daridon@univ-pau.fr.

Funding

CICECO is being funded by Fundação para a Ciência e a Tecnologia through Pest-C/CTM/LA0011/2011.

Notes

The authors declare no competing financial interest.

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