Measurement and prediction of the speed of sound of biodiesel fuels

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HIGHLIGHTS

▶ New data on speed of sound is reported for fatty acid methyl esters.
▶ New data on speed of sound is reported for biodiesel fuels.
▶ A predictive model based on linear mixing rule is proposed.
▶ A successful extension of the model for high pressures is presented.

ARTICLE INFO

ABSTRACT

Speed of sound is an important fuel thermophysical property that directly characterizes the fuel injection and the NO_x emissions in diesel engines especially for injectors activated with pressure. Nevertheless, the experimental data of speed of sound for biodiesel fuels are very scarce in the literature. Thereby, this work aims at measuring the speed of sound for biodiesels fuels and evaluating some predictive models to estimate the speed of sound from the composition of fatty acid methyl esters in the biodiesel.

For that purpose the measurement of speeds of sound for three fatty acid methyl esters and ten biodiesel fuels at atmospheric pressure and temperatures from 288.15 to 343.15 K was done. The ability of two versions of Auerbach's relation, and the ideal mixture mixing rule to describe the speed of sound of biodiesels was evaluated. The results evinced that, with the exception to the original version of Auerbach's relation, the other models studied provide a good estimate of the experimental data. The modified Auerbach's relation displays an overall average relative deviation of 1.64%, and the ideal mixture mixing rules predict the biodiesel speed of sound with only 0.37% of overall deviation. Moreover, the dependency of speed of sound on pressure was also correlated with a linear equation, presenting only an overall deviation of 0.56%. These models can thus be a good tool to estimate the speeds of sound using only information about the biodiesel composition.

1. Introduction

Current researches on fuels have highlighted oftentimes the potential of biodiesel fuels as an alternative to petrodiesel. Whence, in fact, there is an increasing growth of public interest in using biodiesel fuels, especially in countries without fossil resources where these fuels can be a support for domestic economic development and energy sustainability. This trend stems basically from the premise that biodiesel fuels offer many benefits such as renewability (i.e., produced from vegetables oils, animal fats and recycled greases), biodegradability, non-toxicity, lower emissions of greenhouse gases, total miscibility with petrodiesel and especially compatibility with modern diesel engines [1,2].

The combustion of biodiesel fuels, however, can produce higher emissions of nitrogen oxides (NO_x) in comparison with that of petrodiesel. For example, blends of B20 (i.e., 20 vol.% biodiesel with 80 vol.% petrodiesel) increase NO_x emissions by 2–4% while B100 by as much 10%. [3] These emissions are prejudicial to the environment as they form smog, destroy the ozone layer, form nitric acid and irritate the eyes, throat and respiratory system. Nitrogen dioxide (NO_2), and nitrous oxide (N_2O) are about 250 times more threatening to global warming than CO_2 at the same concentration [4–6].

Many approaches have been considered to reduce the NO_x emissions. While some works suggested the use of Exhaust Gas Recirculation with biodiesel [3], others focused on the influences

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of injection and combustion parameters in NO$_x$ emissions such as the injection rate, injection timing, injection duration, injection pressure, start of combustion, in-cylinder gas pressure and temperature and heat release rate [7]. Anyway the dissimilar composition of fuels reflects the differences in physical properties like density, bulk modulus, heat capacity and speed of sound that later affect both the quality of fuel injection system and the diesel engine combustion [2,8–11]. Bio diesel fuels have higher values of density, viscosity, speed of sound, isentropic bulk modulus, cetane number and vapor pressure but lower heating values [8]. Although a higher cetane number would imply a shortened ignition delay period, resulting in relatively reduced NO$_x$ formation [12], a higher isentropic bulk modulus and higher speed of sound would cause an earlier injection of fuel, increasing the emissions of NO$_x$, namely for injectors activated with pressure [10,13,14]. Anyway, the magnitude of these properties is influenced by the structure of the fatty acid alkyl esters that compose biodiesel fuels such as chain length, branching and level of unsaturation [2]. Thus the knowledge of the relationship between the biodiesel properties and its composition in fatty acid methyl esters (FAME) is of great importance. In previous works we have reported new data and proposed predictive methodologies for the description of densities at low and high pressure [15], viscosities [16] and surface tensions [17]. Unfortunately there are only few works on the speed of sound of biodiesel fuels, and even less on the prediction of this property based on the composition of FAME. Ott et al. [18] measured the speed of sound as a function of temperature at 83 kPa for five pure methyl esters and later [19] they reported data for two B100 commercial fuels at 83 kPa. Tat et al. [11] correlated the speed of sound of methyl soyate and ethyl soyate with pressure at 21 °C. Gouw et al. [20] reported the ultrasonic sound velocity of methyl esters at 20 and 40 °C, while Tat and Van Gerpen [13] provided correlated data of the speed of sound for some biodiesel fuels and pure esters at temperatures from 20 °C to 100 °C and pressures from atmospheric to 34.5 MPa. These informations, however, still do not provide an approach for estimating the speed of sound of a biodiesel fuel from its composition.

This work aims to fill this gap by providing the experimental data of speed of sound for three pure methyl esters and ten biodiesel fuels at atmospheric pressure and temperatures from 288.15 to 343.15 K, and then evaluating the predictive ability of Auerbach's relation, and ideal mixture mixing rules for describing the experimental data of biodiesel fuels based on the composition of FAME. An extension of the model to high pressures based on literature data is also attempted.

2. Experimental details

2.1. Biodiesel samples synthesis

The three methyl esters here studied were Methyl Laurate (with 97% of purity from Fluka), Methyl Myristate (with 98% of purity from SAFC) and Methyl Oleate (with 99% from Aldrich). Ten biodiesel fuels were studied: Soy A and GP (Soybean + Rapeseed) were obtained from Portuguese biodiesel producers while the other eight biodiesel samples were synthesized at our laboratory, as described in a previous work [15], by the transesterification reaction of the following vegetal oils with methanol. These are Soybean (S), Rapeseed (R) and Palm (P), and their respective binary and ternary mixtures: Soybean + Rapeseed (SR), Rapeseed + Palm (RP), Soybean + Palm (SP), and Soybean + Rapeseed + Palm (SRP) and Sunflower (Sf).

Detailed compositions of these biodiesels are reported in Table 1. These data were acquired using a Capillary GC of Varian CP-3800 with a FID in a split injection system with a select biodiesel for

FAME column (30 m × 0.32 mm × 0.25 μm). The column temperature was set at120 °C and then programmed to increase up to 250 C at 4 °C/min. The detector and injector were set at 250C. The carrier gas was helium witha flow rate of 2 mL/min. [15].

2.2. Measurement of speed of sound

The ultrasonic pulse echo technique was used to measure the speed of sound of biodiesel fuels from 278.15 to 373.15 Kat atmospheric pressure using the Anton Paar DSA 5000 density and sound velocity analyzer. This instrument is equipped with a density cell and a speed of sound cell combining the known oscillating U-tube method with a highly accurate measurement of speed of sound. It was calibrated against triple-distilled fresh water and air at atmospheric pressure. The calibration was accepted if the measurements were estimated to be within ±2.10^{-3} kg m^{-3} and ±0.01 m/s of the reference values. The measurements were obtained in duplicates, and the standard experimental uncertainty was obtained by dividing the modulus of the repeatability differences by the square of two. The value obtained was 0.23 m/s for 142 repetition points. [21].

3. Models for speed of sound

The description of speed of sound for biodiesel fuels and methyl esters was done by using the Auerbach's relation, an approach previously used with success for ionic liquids [22], alkyl alkanoates and alkyl amines [6,23], and metallic liquids [24], and alternatively using a linear mixing rule.

3.1. Auerbach's model

The Auerbach's model [22,25] is represented by Eq. (1), where u is the speed of sound in m/s, γ is the surface tension in N m^{-1}and ρ is the density in kg m^{-3}. Since this equation requires the prior knowledge of densities and surface tensions, this work uses the data reported in our previous works [15,17].

$$u = \left(\frac{\gamma}{6.33 \times 10^{-10} \cdot \rho} \right)^{\frac{2}{3}} \tag{1}$$

This work also considered a modified version of Auerbach's model, in order to achieve a better description of the experimental speed of sound data for biodiesel fuels, by relaxing the value of the constant $c1$ of the following equation:

$$u = \left(\frac{\gamma}{6.33 \times 10^{-10} \cdot \rho} \right)^{\left(\frac{2}{3} \cdot c1\right)} \tag{2}$$

3.2. Ideal mixture mixing rules

As biodiesel fuels are a mixture of FAME of similar molecular weight, their speeds of sound can be estimated using a mixing rule assuming an ideal mixture behavior. This approach, described by Eq. (3), will be here used to describe the speed of sound of biodiesels, where u_{BD} is the speed of sound of biodiesel in m/s, x_i is the molar composition and u_i is the speed of sound of individual FAME in m/s.

$$u_{BD} = \sum_i x_i u_i \tag{3}$$

Due to lack of experimental data of speed of sound for some minoritory FAME compounds, such as C10:0, C16:1, C20:0, C20:1, C22:0, C22:1 and C24:0,to predict the speed of sound of biodiesel fuels containing these compounds, in this work the

Table 1
Compositions of the Biodiesels studied, in mass percentage.

FAME	S	R	P	SR	PR	SP	SRP	Sf	GP	Soy A
C10:0	0	0.01	0.03	0	0.02	0.01	0.01	0	0	0
C12:0	0	0.04	0.24	0.03	0.20	0.18	0.14	0.02	0.02	0
C14:0	0.07	0.07	0.57	0.09	0.54	0.01	0.38	0.07	0.13	0
C16:0	10.76	5.22	42.45	8.90	23.09	25.56	18.97	6.40	10.57	17.04
C16:1	0.07	0.20	0.13	0.15	0.17	0.11	0.14	0.09	0.13	0
C18:0	3.94	1.62	4.02	2.76	3.02	4.04	3.28	4.22	2.66	3.73
C18:1	22.96	62.11	41.92	41.82	52.92	33.13	42.51	23.90	41.05	28.63
C18:2	53.53	21.07	9.80	37.51	15.47	31.72	27.93	64.16	36.67	50.45
C18:3	7.02	6.95	0.09	7.02	3.08	3.58	4.66	0.12	7.10	0
C20:0	0.38	0.60	0.36	0.46	0.49	0.39	0.45	0.03	0.44	0
C20:1	0.23	1.35	0.15	0.68	0.67	0.20	0.52	0.15	0.67	0
C22:0	0.80	0.35	0.09	0.46	0.24	0.32	0.33	0.76	0.45	0
C22:1	0.24	0.19	0.00	0.12	0.09	0.12	0.14	0.08	0.12	0
C24:0	0	0.22	0.15	0	0	0.63	0.53	0	0	0

pseudo-component concept was adopted by adding C10:0 to C12:0, C16:1 to C16:0 and C20:0, C22:0 and C24:0 to C18:0 and C20:1 and C22:1 to C18:3. In this work the experimental speed of sound for the methyl esters used, with their respective purities, was that reported by Tat and Van Garpen [13] except for C14:0 measured in this work. This methyl ester has 98% of purity. The nomenclature for esters here adopted is based on the fatty acid chain length. So Cx:y ester represents the methyl ester of fatty acid with x carbons and y unsaturations.

3.3. Model evaluation

To evaluate the predictive capacity of the models aforementioned, the relative deviations (RDs) between the predicted and experimental data of speed of sound were estimated according to Eq. (4).

$$RD(\%) = \frac{u_{calc_i} - u_{exp_i}}{u_{exp_i}} \times 100 \qquad (4)$$

The average relative deviation (ARD) was calculated as a summation of the modulus of RD over N_p experimental data points. The overall average relative deviation (OARD) was calculated by Eq. (5), where N_s is the number of systems studied.

$$OARD(\%) = \frac{\sum_n ARD}{N_s} \qquad (5)$$

4. Results and discussion

The experimental data of speed of sound for three pure methyl esters and ten biodiesels here studied are reported in Tables 2 and 3. The experimental data of speed of sound for the methyl esters

Table 2
Experimental Speed of sound, in m/s, for methyl esters.

T (K)	Methyl laurate	Methyl myristate	Methyl oleate
288.15	1369.55	NM	1427.25
293.15	1350.85	NM	1409.44
298.15	1332.26	1353.19	1391.68
303.15	1313.78	1334.98	1374.06
308.15	1295.44	1316.91	1356.62
313.15	1277.24	1298.96	1339.31
318.15	1259.25	1281.21	1322.15
323.15	1241.39	1263.57	1305.06
328.15	1223.68	1246.11	1288.19
333.15	1206.08	1228.73	1271.39
338.15	1188.39	1211.48	1254.72
343.15	1171.08	1194.32	1238.21

NM – No measurement.

here measured were compared to those reported in the literature. The relative deviations are shown in Fig. 1, where it is seen that a very good agreement is obtained with the deviations within ±1% of literature data. Very small differences in the speed of sound are observed between the various biodiesels studied. Both the magnitude of the speed of sound and its temperature dependency vary less than 1% between the fluids, unlike what was previously observed for other properties [9,17,21]. In spite of the similarities it can be observed, nevertheless, that the increase in concentration of the saturation level of the compounds decreases the speed of sound. The palm biodiesel and the mixtures containing palm present the lower speeds of sound, while the sunflower has the largest speed of sound measured in this work.

The models described above were applied to the description of the data here measured. The results reported in Table 4 show that the original version of Auerbach's equation fails to provide a good description of the experimental data. Aiming at enhancing the description of the data the value of the model parameter was modified. To develop the modified Auerbach's relation, four methyl esters (methyl palmitate, methyl oleate, methyl stearate and methyl linoleate) reported by Ott et al. [18] and three biodiesels (S, SR and SRP) were used as training set to adjust the value of $c1$, while the other methyl esters and biodiesels here measured were used as validation set. A value for $c1$ of 0.987 was obtained that provided an OARD of 1.34% for the training set and of 1.42% for the validation set in the temperature range 283–373 K. The behavior of the modified Auerbach's model for both set of compounds can be seen separately in Figs. 2a and b where the deviation between the predicted and experimental data is within ±4%. A deficient temperature dependency of the model is highlighted in these figures.

The average relative deviations (ARD) for the ten biodiesels here studied and eleven other biodiesels previously reported in the literature are presented in Table 4 for the various models investigated. Here it can be seen that the ideal mixture mixing rules is seen to be the more appropriate model for describing the speeds of sound for biodiesel fuels, presenting only an OARD of 0.37% for 21 biodiesel fuels here studied and the individual value of ARD was almost of the same magnitude for all biodiesels in comparison with 9.13% and 1.64% of the original Auerbach's equation and the modified Auerbach's model respectively.

The behavior of predictions of speed of sound for biodiesel fuels are shown in Fig. 3. It is possible to observe again that the ideal mixture mixing rules is the most appropriate model for describing the experimental data, allowing a good prediction of their temperature dependency.

To estimate the speed of sounds for biodiesel fuels at high pressures, this work used the experimental data of speed of sound reported by Tat and Van Gerpen [13] to develop a correlation. The experimental data displays very similar pressures dependencies

Table 3
Experimental speed of sound, in m/s, for methylic biodiesel.

T (K)	S	R	P	SR	RP	SP	SRP	Sf	GP	Soy A
288.15	1430.23	1430.79	1420.04	1430.66	1424.52	1424.92	1426.90	1432.34	1428.88	1428.53
293.15	1412.39	1412.26	1401.86	1412.77	1406.55	1406.86	1408.97	1414.56	1410.97	1410.73
298.15	1394.54	1394.11	1383.88	1394.95	1388.65	1388.94	1391.11	1396.53	1393.10	1392.90
303.15	1376.86	1376.22	1366.08	1377.27	1370.89	1371.17	1373.36	1378.54	1375.39	1375.17
308.15	1359.20	1358.40	1348.38	1359.74	1353.34	1353.61	1355.80	1360.74	1357.86	1357.63
313.15	1341.80	1340.88	1331.14	1342.37	1335.91	1336.17	1338.37	1343.04	1340.47	1340.16
318.15	1324.55	1323.55	1313.88	1325.16	1318.60	1318.86	1321.11	1325.80	1323.21	1322.90
323.15	1307.37	1306.31	1296.69	1308.07	1301.48	1301.73	1303.97	1308.63	1306.07	1305.75
328.15	1290.35	1289.26	1279.72	1291.15	1284.49	1284.74	1287.00	1291.53	1289.12	1288.79
333.15	1273.43	1272.30	1262.94	1274.31	1267.62	1267.86	1270.13	1274.60	1272.26	1271.91
338.15	1256.69	1255.59	1246.18	1257.61	1250.85	1251.09	1253.40	1257.76	1255.52	1255.17
343.15	1240.02	1239.05	1229.66	1241.09	1234.28	1234.51	1236.84	1241.05	1238.99	1238.53

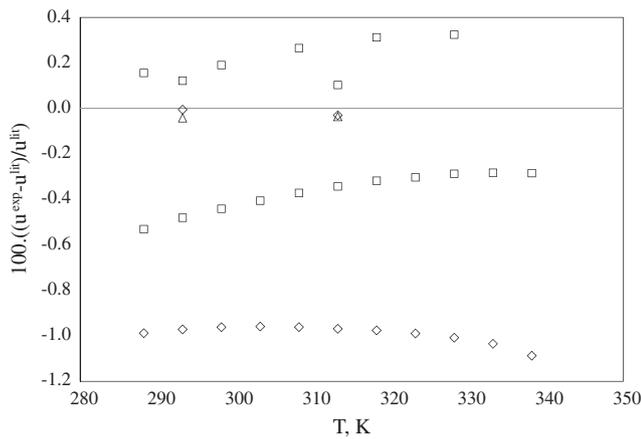**Fig. 1.** Relative deviations of the speed of sound of three methyl esters here studied ◇ Methyl Laurate, △ Methyl Myristate and □ Methyl Oleate [13,18,20].
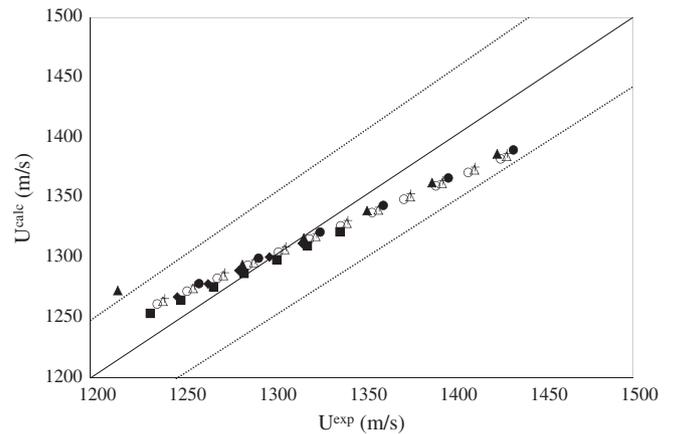

Fig. 2a. Predicted vs. experimental speed of sound of the training set for modified Auerbach's model. ■ Methyl palmitate, ♦ Methyl stearate, ▲ Methyl oleate, ● Methyl linoleate, △ S, + SR, ○ SRP and …±4%.

Table 4
Average relative deviations (ARD) of speed of sound for biodiesel fuels using the models here studied.

Compound	ARD, %		
	Auerbach original	Modified auerbach	Ideal mixture
BD–A [19]	8.13	1.91	0.18
BD–B [19]	8.40	1.80	0.23
BD–JC [1]	7.51	2.10	0.11
Methyl Soy ester [13]	7.80	1.91	0.09
Methyl Canola [13]	8.22	1.66	0.20
Methyl Tallow [13]	8.02	1.95	0.76
Methyl Lard [13]	8.49	1.64	0.24
Methyl oxidized soy [13]	11.35	1.89	1.07
Methyl hydrogenated soy [13]	7.85	1.92	0.39
S	9.28	1.49	0.34
Soy A	8.00	1.18	0.29
R	10.53	1.45	0.33
P	11.70	1.47	0.46
Sf	9.84	1.43	0.33
SP	10.48	1.19	0.36
SR	8.95	1.45	0.25
PR	11.33	1.47	0.46
SRP	9.86	1.47	0.38
GP	9.84	1.20	0.37
NIST SRM 2772 B100 [27]	7.91	1.99	0.34
NIST SRM 2773 B100 [27]	8.21	1.78	0.53
OARD, %	9.13	1.64	0.37

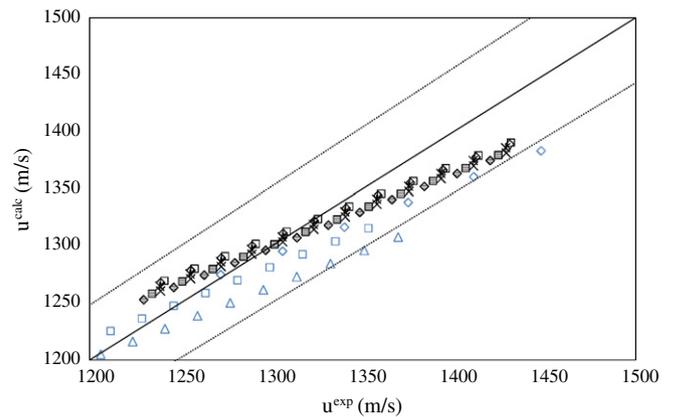Where BD–A and BD–B are two types of commercial B100 and BD–JC is the *Jatropha curcas* Biodiesel.

Fig. 2b. Predicted vs. experimental speed of sound of the validation set for the modified Auerbach's model. △ Methyl Laurate, □ Methyl Myristate, ◇ Methyl oleate, ✴ soy A, ◇ R, ◖ P, □ Sf, × GP, ▬ SP and …±4%.

for the speeds of sound observed for the pure FAME's and for the biodiesel fuels, that are linear in the range of pressures (0–35 MPa) studied by Tat and Van Gerpen [13]. For the same pressure range, a linear behavior is also observed for the experimental data reported by Payri et al. [26] It should thus be possible to describe the pressure dependency of the speed of sound up to 40 MPa by
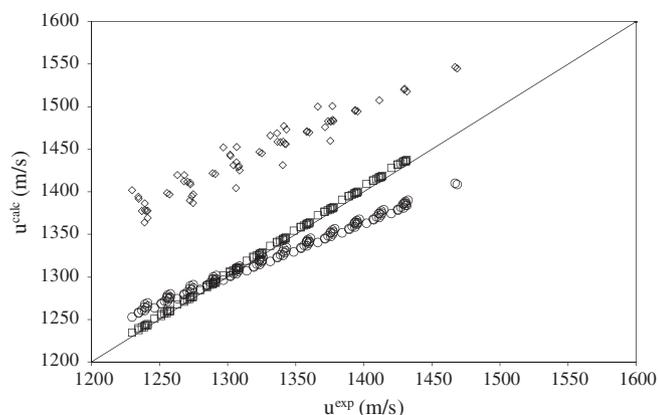
$$u = u_0 + aP \tag{6}$$

Fig. 3. Predictive ability of the three models evaluated in describing the experimental data of speed of sound for the biodiesel fuels here studied: ◇ Auerbach original, ○ Modified Auerbach and □ Ideal mixture mixing rules.

Table 5
Average relative deviations (ARD) of speed of sound for methyl esters and biodiesel fuels at high pressure [13,26].

Compounds	ARD, %	
	Training set	Validation set
Methyl Laurate	0.46	
Methyl Palmitate	0.38	
Methyl Stearate	0.35	
Methyl Oleate	0.34	
Methyl Linoleate	0.34	
Methyl Linolenate	0.35	
Methyl Soy Ester		0.35
Methyl Canola		0.34
Methyl Tallow		0.37
Methyl Lard		0.33
Methyl Oxidized Soy		0.35
Methyl Hydrogenated soy		0.24
Rapeseed Methyl Ester		0.82
OARD, %	0.37	0.56

Where u is the speed of sound in m/s, u_0 is the speed of sound at atmospheric pressure, a is the pressure gradient and P is the pressure in MPa.

To develop a high pressure correlation for the speed of sound of biodiesel fuels the six FAMEs, C12:0, C16:0, C18:0, C18:1, C18:2 and C18:3 reported by Tat and Van Gerpen. [13], were used as the training set while the other six biodiesels as the validation set. After fitting the Eq. (6) to the experimental data of the training set, the numerical value of the pressure gradient was obtained (a = 4.53 m/s MPa^{-1}). This correlation of speed of sound provided an OARD of 0.37% for the training set and 0.56% for the validation set as detailed in Table 5. The validity of these correlations is limited to the pressures below 40 MPa and should not be extrapolated to higher pressures. As shown by Payri et al. [26] for higher pressures the pressure dependency of the speed of sound is no longer linear. Unfortunately at present the data available precludes the development of a correlation for higher pressures.

5. Conclusions

The experimental data of speed of sound for three pure methyl esters and ten biodiesel fuels were measured at temperatures from 288 to 343 K and at atmospheric pressure. These data were then used, along with other literature data, to evaluate the capacity of two versions of Auerbach's relation, and ideal mixture mixing rules to predict the speed of sound of biodiesel fuels from the knowledge of their composition. For all biodiesel studied, the overall average relative deviation (OARD)value obtained for these models were 1.64 and 0.37% respectively for the modified Auerbach model and ideal mixture mixing rules. A correlation for estimating the speed of sounds for biodiesels at high pressures was also developed. It is valid up to 40 MPa and provides an OARD of 0.56% for seven biodiesels tested. Therefore when the measurement of speed of sound is impractical for any biodiesel, these models can be a useful tool for predicting the speed of sound in a wide range of temperatures and pressures provided that the composition of FAME is known.

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