Speed of sound in biodiesel produced by low power ultrasound

P A Oliveira, R M B Silva, G C Morais, A V Alvarenga and R P B Costa-Felix
Laboratory of Ultrasound (Labus), National Institute of Metrology, Quality and Technology (Inmetro), Av. Nossa Sra. das Graças 50, Duque de Caxias, RJ, Brazil, ZIP 25250-020

passuncaooliveira@gmail.com; rpfelix@inmetro.gov.br

Abstract. The quality control of the biodiesel produced is an important issue to be addressed for every manufacturer or retailer. The speed of sound is a property that has an influence on the quality of the produced fuel. This work presents the evaluation about the speed of sound in biodiesel produced with the aid of low power ultrasound in the frequencies of 1 MHz and 3 MHz. The speed of sound was measured by pulse-echo technique. The ultrasonic frequency used during reaction affects the speed of sound in biodiesel. The larger expanded uncertainty for adjusted curve was 4.9 m.s⁻¹.

1. Introduction
The search for alternative energy sources has been motivated due to the limitation and exhaustion prediction of fossil fuels, besides environmental concerns. Thus, the production and use of biodiesel became attractive for being a renewable fuel.

One of the limiting factors for the large-scale commercialization of biodiesel is the relative high cost of production compared to fossil fuels [1]. Hence, improvements in biodiesel production processes are necessary to provide costs reduction. Ultrasound has been used in biodiesel production for this purpose [2][5].

It is known that the quality control of the biodiesel produced is extremely important. In the injection process, fuel quality directly affects engine efficiency. Some properties such as viscosity and density strongly influence the quality of the produced fuel [6]. The speed of sound is way to assess these parameters and consequently the quality of the fuel. Thus, the determination of the speed of sound in biodiesel is relevant [7].

However, few studies available in the literature determine the speed of sound in biodiesel [6][10]. Commonly, the cited authors studied the biodiesel produced from the conventional method (mechanical stirring and heating) and rarely consider the metrological aspects.

Therefore, this paper aims to evaluate the speed of sound for biodiesel produced by ultrasound-assisted through low-power, in the frequencies of 1 MHz and 3 MHz. Throughout the study the calculation of uncertainties of measurements was carried.
2. Materials and methods

2.1. Biodiesel synthesis
The biodiesel was synthesized by the transesterification reaction of soybean oil (Liza®) with methanol (purity 99.8%) using molar ratio of alcohol/oil of 6:1 and potassium hydroxide as a catalyst (1.5%). The reaction was carried out in a mini-reactor using two ultrasound generator devices models 4144 and 4134 (Sonomed, Brasil), operating in frequencies of 1 MHz and 3 MHz with nominal power of 7.2 W. The reaction was performed without the aid of mechanical stirring or heating bath, and its total time was 40 minutes.

2.2. Measuring the speed of sound
The technique described in [11] was used in the measurement of the biodiesel speed of sound. The measurement system is shown in figure 1, and is composed by a thermal bath (a) a cell with biodiesel (b), and an ultrasonic transducer (c) of 1 MHz and 12.7 mm in diameter model A303S (NDT-Panametrics, Olympus Corporation, Japan) [12]. The ultrasonic transducer acted as transmitter and receiver, and it was excited through a wave generator (d) model 33250A (Agilent Technologies, CA, USA). The acquired signals were digitized by an oscilloscope (e) model DSO-X 3012A (Agilent Technologies, CA, USA) and transferred to a computer (f) through a program developed in LabView (National Instruments, TX, USA).

![Figure 1. Experimental Setup.](image)

The speed of sound in each temperature, for each repetition, was determined through the measurement of the time that the ultrasonic signal took to cross the sample and return to the same transducer (time of flight). The temperature of bath was set initially at 20 °C and increases up to 50 °C. It was monitored during the measurement using a temperature measuring device (g) model 34970A (Agilent Technologies, CA). The measurements were performed under repeatability conditions, with five measurements for each biodiesel sample [13][14].

According to Oliveira et al [11], the speed of sound in vegetable oils as a function of temperature is given by a linear function. Thus, after determining the biodiesel propagation velocity at each point, adjusted curves were established ($v_{bioa}$) based on equation (1).

$$v_{bioa} = (a + b \cdot T) + \delta$$

in which $a$ and $b$ are constants obtained in each repetition (contributing to uncertainty of type A), and $T$ is the temperature in °C. The type B uncertainty, derived from the calibration certificate of the thermometer, was used in the uncertainty budget. Since $\delta$ is a term that has been inserted in the
equation in order to take into account the uncertainty of each measured velocity point, its value is equal to zero but it has an associated uncertainty value of 0.65 m.s\(^{-1}\).

The expanded uncertainty was calculated according to the Guide of the Expression of Uncertainty in Measurements [15], by a coverage factor \(k\) that takes into account a t-distribution with coverage probability of 0.95.

2.3. Normalized error
The results of speed of sound of the produced biodiesel were statistically compared by calculating the normalized error [16], according to equation (2).

\[
E_n = \frac{|x_{1\text{MHz}} - x_{3\text{MHz}}|}{\sqrt{U_{1\text{MHz}}^2 + U_{3\text{MHz}}^2}}
\]

(2)

in which \(E_n\) is the normalized error; \(x_{1\text{MHz}}\) is the speed of sound in biodiesel produced at 1 MHz; \(x_{3\text{MHz}}\) is the speed of sound in biodiesel produced at 3 MHz; \(U_{1\text{MHz}}\) and \(U_{3\text{MHz}}\) are the respective the expanded uncertainties \((p = 0.95)\).

The normalized error tests the compatibility of the results; its evaluation criterion is given by: \(|E_n| \leq 1.0\) the biodiesel speeds of sound are metrologically equivalent; \(|E_n| > 1.0\) the biodiesel speeds of sound are not statistically equivalent.

3. Results and discussion
The results of speed of sound obtained from the five replicates for soybean biodiesel in 1 MHz and 3 MHz are shown in figures 2 and 3, respectively.

![Figure 2](image-url) **Figure 2.** Relationship between speed of sound and temperature for biodiesel produced at 1 MHz.

![Figure 3](image-url) **Figure 3.** Relationship between speed of sound and temperature for biodiesel produced at 3 MHz.

One can observe that the relation between the speed of sound in biodiesel and the temperature tends to be linear for the studied temperature range (figure 2 and 3). Besides, for all the five repetitions, the coefficients of determination \((R^2)\) are close to 1. This corroborates the linear relation between the variables studied.

Adjusted curves were established based on linear regression data from the five measurements performed for each biodiesel, and the expanded uncertainty was estimated, as shown in tables 1 and 2.
Table 1. Expanded uncertainty ($U_{1MHz}$) of the adjusted curve for speed of sound for biodiesel produced with 1 MHz ultrasonic frequency.

| $T$ [°C] | $v_{bio_a}$ [m.s$^{-1}$] | $k$ [p=0.95] | $U_{1MHz}$ [m.s$^{-1}$] |
|----------|--------------------------|--------------|-------------------------|
| 20       | 1450.8                   | 2.36         | 4.3                     |
| 25       | 1435.1                   | 2.36         | 4.4                     |
| 30       | 1419.5                   | 2.26         | 4.3                     |
| 35       | 1403.8                   | 2.23         | 4.4                     |
| 40       | 1388.2                   | 2.18         | 4.5                     |
| 45       | 1372.5                   | 2.14         | 4.6                     |
| 50       | 1356.9                   | 2.12         | 4.7                     |

Table 2. Expanded uncertainty ($U_{3MHz}$) of the adjusted curve for speed of sound for biodiesel produced with 3 MHz ultrasonic frequency.

| $T$ [°C] | $v_{bio_a}$ [m.s$^{-1}$] | $k$ [p=0.95] | $U_{3MHz}$ [m.s$^{-1}$] |
|----------|--------------------------|--------------|-------------------------|
| 20       | 1435.0                   | 2.36         | 4.4                     |
| 25       | 1419.9                   | 2.36         | 4.6                     |
| 30       | 1404.7                   | 2.26         | 4.5                     |
| 35       | 1389.6                   | 2.23         | 4.6                     |
| 40       | 1374.5                   | 2.18         | 4.7                     |
| 45       | 1359.4                   | 2.14         | 4.8                     |
| 50       | 1344.2                   | 2.11         | 4.9                     |

The graphical representation of tables 1 and 2 is shown figure 4.

![Figure 4](image-url)

Figure 4. Adjusted curves with expanded uncertainty for each biodiesel studied.

Then, from the data in tables 1 and 2, the normalized error was calculated to determine if the speeds of sound biodiesel samples produced from different frequencies are statistically equivalent. The results are presented in table 3.
Table 3. Normalized error for evaluation between biodiesel produced by different frequencies.

| $T$ [°C] | $E_a$   |
|---------|---------|
| 20      | 2.56    |
| 25      | 2.40    |
| 30      | 2.38    |
| 35      | 2.23    |
| 40      | 2.10    |
| 45      | 1.98    |
| 50      | 1.86    |

Analyzing table 3, it is possible to note that speed of sound in biodiesel produced by different ultrasonic frequencies are not statistically equivalent, since the normalized error was greater than 1. The frequency of the ultrasonic waves is one of the factors that lead to the occurrence of the cavitation bubbles. Different frequencies form cavitation bubbles with different diameters, which may influence the emulsification of the reaction and consequently the biodiesel produced.

4. Conclusions

The speed of sound for two biodiesels produced from different frequencies of ultrasonic irradiation was measured, varying the temperature from 20 °C to 50 °C.

For biodiesel studied here, the speed of sound decreases with increasing temperature and this relationship is well described by a linear function, what is not the case for other biological tissues. Thus, the results agree with the behavior of speed of sound of data available in the literature for conventional biodiesel. The correlation coefficient and the low uncertainty disclose the quality of the measurements, reinforcing the importance of metrological aspects for those measurements.

The first set of results show that the ultrasound frequency used during the biodiesel production affects directly the speed of sound in the final product. However, to determine which frequency is most appropriate for the biodiesel production by ultrasound, analysis of the conversion rate of all the reactions performed is necessary, as well a comparison with the properties of the conventional biodiesel.

The difference in the speed of sound in the produced biodiesels is an evidence that they have dissimilar physical chemical properties. As, within this research, biodiesel was produced with the aid of different ultrasound frequencies, one can infer directly that ultrasonic parameters play a key role in the reaction.

References

[1] Glisic S B, Pajnik J M and Orlovic A M 2016 *Applied Energy*. v 70 p 176-185.
[2] Than L T, Okitsu K, Maesa Y and Bandiw H 2014 *Ultrasonics Sonoch* . v 21(2) p 467-441.
[3] Moholkar V S, Choudhury H A and Srivastava P 2014 *AIChE Journal*. v 60(5) p 1572-1581.
[4] Ferreira E, Vicente J, Silva R C D, Nele M, Nunes E and Costa-Felix R P B 2014 *Polymer Testing*. 33 pp 16-20.
[5] Oliveira J V, Oliveira D, Treichel H, Rosa C D, Batistella L, Popiolki A S and Trentin C M 2015 *Bioprocess Biosyst. Eng*. v 38 p 437-448.
[6] Lopes A F G, Talavera-Prieto M C, Ferreira A G M, Santos J B, Santos M J and Portugal A T G 2014 *Fuel*. v 116 p 242–254.
[7] Maggi L E, Silva C E R, Alvarenga A V and Costa-Felix R P B 2011 *Journal of Physics. Conference Series*. 279 pp 012029.
[8] Freitas S V D, Paredes M L L, Daridon J, Lima A S and Coutinho J A P 2013 *Fuel*. v 103 p 1018-1022.
[9] Ndiaye H I, Habrioux M, Coutinho J A P, Paredes M L L and Daridon J L 2013 J Chem Eng Data. 58 p 1371–1377.
[10] Tat M E and Gerpen J H V 2003 J Am Oil Chem Soc. 80 p 1249-1256.
[11] Oliveira P A, Baësso R M, Morais G C, Alvarenga AV and Costa-Félix R P B 2016 J. Phys.: Conf. Ser. 733 pp 012040.
[12] Silva C E R, Alvarenga A V and Costa-Félix R P B 2011 J. Phys.: Conf. Ser. 279 pp 012025.
[13] Costa-Félix R P B 2006 Measurement. 39 pp 169-175.
[14] Costa-Félix R P B and Machado J C 2015 Measurement. 69 pp 146-154.
[15] BIPM, JCGM:100 2008 Paris.
[16] Steele A G and Douglas R J 2006 Metrologia. 43(4) p S235-S243.
[17] Abrunhosa V M, Soares C P, Possidonio A C B, Alvarenga A V, Costa-Félix R P B, Costa M L P S and Mermelstein C S 2014 Ultrasound in Medicine & Biology. 40 pp 504-512.
[18] Petrella L I, Maggi L E, Souza R M, Alvarenga A V and Costa-Félix R P B 2014 Ultrasonics. 54 pp 1476-1479.

Acknowledgements
Research reported in this paper has been partially supported by the Carlos Chagas Filho Research Support Foundation (Faperj, grant number E-26/201.563/2014), by the National Council for Scientific and Technological Development (CNPq, grant number 310.392/2014-4) and Pronametro/Inmetro.