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In-pipe coconut water rheological characterization with ultrasonic waves

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ABSTRACT

We propose in this paper to use a simple and robust experimental protocol based on longitudinal ultrasonic velocity measurement in order to evaluate the viscosity of coconut water in a cylindrical stainless-steel pipe. Seven samples with Soluble Solids Content (SSC) ranging from 6.7 to 44.2°Brix were studied using conventional Couette viscometry and high-frequency ultrasonic methods. Calibration laws linking the ultrasonic velocity measured at 5 MHz to the shear viscosity and to the SSC are proposed. These laws are in very good agreement with previous measurements carried out several years ago using a plane 25 MHz transducer directly introduced into the coconut water with SSC of up to 60°Brix.

1. Introduction

Among the important rheological parameters, viscosity is certainly the most investigated because it reflects the flowing capacity of the material and thus provides a lot of data on the microstructure (Ferry, 1961). In particular, for sweet foods and beverages, the sugar content and sugar composition (glucose, fructose, sucrose …) is directly related to the viscosity (Telis et al., 2007). Moreover, this data is fundamental in many industrial applications as its understanding leads to control and optimization of manufacturing processes (Povey and Mason, 1998). Many experimental methods dedicated to viscosity evaluation have been developed. However, the most promising are the low-power ultrasonic approaches because they provide to a non-destructive measurement method and can be carried out through thick and opaque walls. The ultrasonic measurement is therefore possible through pipes during the filtration, concentration and processing of juices. In literature, many authors have described various ultrasonic methods dedicated to the characterization of fluids in food engineering (Contreras et al., 1992; Kuo et al., 2008; McClements and Gunasekaran, 1997; Resa et al., 2004; Saggin and Coupland, 2002). Most of the time, the measurement is carried out ex-situ. This means that a sample is extracted from the production line and analyzed in a specific device. It can be noticed that similar recent approaches are used in biomedical domain for blood analysis (Voleisis et al., 2017). In contrast, in other sectors such as the petroleum industry, there are many approaches dedicated to direct measurement in pipeline using either propagating waves in the oil or guided waves in the walls of the pipeline (Kazys and Rekuviene, 2011; Ma et al., 2007; Pavlakovic, 1998; Rose, 1998).

In the case of coconut water, to our knowledge, no, or only a few, scientific papers are dedicated to the link between ultrasonic parameters and viscosity or SSC. For instance, recently, Samuel (Samuel et al., 2015), described an ex-situ ultrasonic measurement using a commercial ultrasonic interferometer to evaluate the effect of laser exposure on a coconut water concentration. In fact, ultrasound are generally used to emulsify mixtures (Ramisetty et al., 2015), to treat coconut water from a chemical point of view (Rojas et al., 2017) or to extract components (Rodrigues and Pinto, 2007), but in these cases the ultrasonic intensity is high and it is not possible to consider these methods as non-destructive. Furthermore, viscosity is not measured. A comprehensive review of such approaches is given in the recent paper of Paniwnyk (2017). Finally, concerning viscosity measured with a rheometer as a function of SSC, one may refer to the work of Manjunatha (Manjunatha and

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Raju, 2013).

In a previous study (Laux et al., 2014), we published measurements of viscosity with the ultrasonic approach ex-situ using a 25 MHz ultrasonic transducer. Thanks to the measurement of longitudinal ultrasonic velocity and attenuation, the link between the longitudinal viscosity obtained by ultrasound and the shear viscosity (measured by rheometer) was established. In conclusion, our proposal was to further adapt the ultrasonic measurements to cylindrical geometry in order to carry out in-line measurements directly on pipes. Therefore, in this new communication we first described an experimental protocol dedicated to longitudinal measurements in cylindrical tubes. The specific signal processing based on a double Fast Fourier's Transform (FFT) was detailed and its robustness was tested on simulated signals. New results obtained on seven coconut water solutions with SSC up to 44° Brix were then presented. We only focused our attention on longitudinal velocity and on the relationship between this velocity, the shear viscosity measured with a Couette rheometer and the SSC.

2. Materials and methods

2.1. Samples

Seven samples were prepared as follows. Coconut water was extracted from 15 immature coconut fruits harvested from the Green Dwarf variety of Thailand (Cock Brand, Thailand). Immature coconut water (CW) extracts were collected and the juice was dispatched in 1 L glass bottles prior to immediate freezing at −50 °C and stored at −18 °C until processing. CW concentration was carried out using a rotary evaporator under controlled pressure conditions. Soluble solids content (SSC) was measured on native and concentrated samples using a digital hand-held PAL refractometer 0–85° Brix (ATAGO Corp., LTD, Japan). CW samples were concentrated 5–10-fold, resulting in Brix values from 6.7 to 44.2°Brix. After concentration, the samples were stored at −18 °C in 20 mL sterile containers. Prior to viscosity measurement, the samples were thawed at 20 °C.

2.2. Flow tests

Shear viscosity was measured using the same protocol as described in our previous study and measurements were carried out using a Physica MCR301 Rheometer (Anton Paar© France, Courtaboeuf, France). A Couette flow measuring cell DG27/T2000/SS was again used. All measurements were carried out at 20 ± 0.1 °C using a Peltier system to control the temperature and a fluid circulator Viscotherm VT 2 controlled directly from the Physica MCR. After 5 min of thermal stabilization, each 11 mL sample was submitted to a flow test in the 10 to 400 s^{-1} shear rate range. The rheological behavior identified was Newtonian and consequently, the viscosity was directly calculated from the ratio between the shear stress and the shear rate. For the seven samples the viscosity ranges between 1.4 and 10.27 mPa s.

2.3. Ultrasonic approach

2.3.1. Ultrasonic high-frequency longitudinal waves in cylindrical pipes

Measurement of the sound velocity in a cavity is a widely investigated subject in literature. Among the works referring to such measurements in liquids in rectangular or cylindrical geometry, we can cite the works of Sinha (Sinha and Kaduchak, 2001). For measurements in gases a large study was carried out by Rosenkrantz (Rosenkrantz, 2007; Rosenkrantz et al., 2009). In cubic cavities it is possible to consider that the ultrasonic waves are plane. Thus, in order to evaluate the velocity of propagation, it is sufficient to divide the distance traveled by the travel time also called time of flight. For cylindrical shells the problems is more complex and is theoretically well known (Rose, 1998). But, if the frequency is high enough to consider that the wavelength is small with respect to the radius of curvature of the cylindrical pipe, it is possible to measure the celerity in a simple way as for plane waves in infinite medium. This point is clearly detailed by the authors previously cited. Thus, the elementary relationship "distance = velocity x time of flight" remains valid. Concerning the measurement of the attenuation, Sinha (Sinha and Kaduchak, 2001) describes two approaches: one in the temporal domain and one using a frequential approach. In order to correct diffraction effects and to take into account the transfer function of the experimental device, a pre-calibration with a known liquid is necessary (Blahova, 2010). As we preferred to avoid such a calibration, we only focused our attention on the ultrasonic velocity evaluation.

2.3.2. Ultrasonic echograms simulation

In order to simulate the ultrasonic signal after propagation in a small cell, the "easier" method is to use the convolution between a typical echo pattern and a reflectivity. This method is generally used in seismic research for blind deconvolution methods testing. In a first approximation the echo pattern can be defined as a sine function multiplied by a Gaussian trend. The reflectivity is defined as a succession of Dirac spikes regularly spaced in temporal domain according to the time of flight. An exponential decay can be added in order to simulate damping due to viscosity. Finally, white noise can also be superimposed. This procedure is illustrated in Fig. 1. The regular space between the echoes is equal to $2d/C_L$ whereby C_L represents the longitudinal ultrasonic velocity in the fluid and d the cell size.

2.3.3. Experimental method

A classical broadband plane Sonaxis© transducer (central frequency 5 MHz – Bandpass 2 MHz) excited with an Olympus© 5800 pulse generator was used and coupled to a stainless-steel tube filled with coconut water. The coupling between the transducer and the tube was ensured using a specific stainless-steel adapter in order to convert plane waves into a cylindrical ultrasonic field (see Fig. 2). The use of this low-cost homemade adapter enabled optimization of the coupling between the transducer and the tube. Secondly, if the diameter of the tube is modified the adapter can easily be reworked. Honey was introduced at the interfaces (transducer/adapter) and (adapter/pipe) and acted as coupling fluid. It would also have been possible to work with a specific ultrasonic

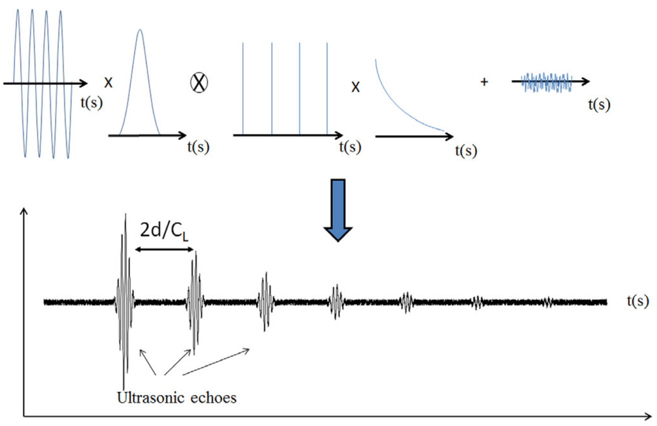

Fig. 1. Numerical generation of ultrasonic echoes.

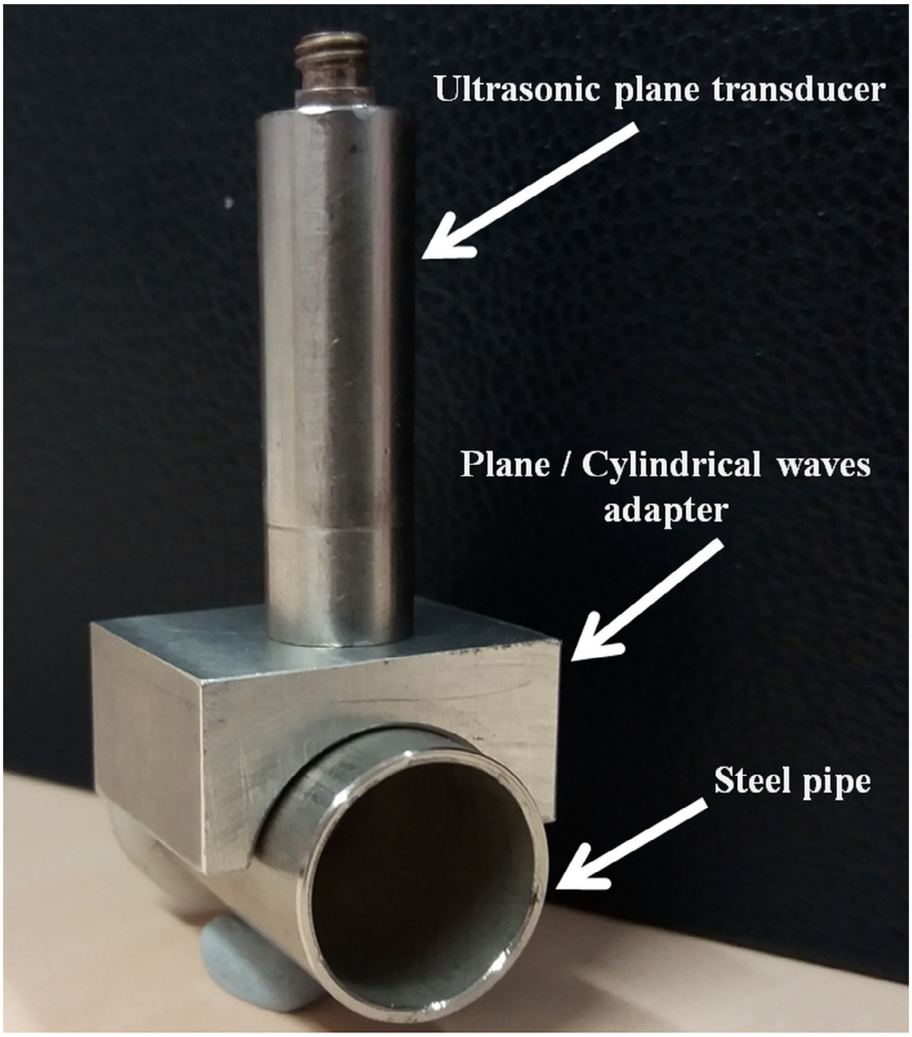

Fig. 2. Photography of the plane transducer and its cylindrical adapter.

transducer dedicated to the emission of cylindrical waves. However, in this case, one would have to buy a specific transducer for each diameter leading to higher experiment costs. The echogram, corresponding to successive passages through coconut water were displayed on a LT374M Lecroy© oscilloscope and acquired by a computer via an USB/GPIB (National Instruments©) connection. In this experimental protocol there is no coconut water flow in the pipe. In a real-life industrial process water flow exists. However, the influence of this flow velocity can be ignored as it is very small compared to the ultrasonic velocity in coconut water.

2.3.4. *Signal processing*

Generally, in order to evaluate the ultrasonic velocity, the authors directly evaluate the time of flight of the ultrasonic signal in the temporal domain. For instance, the position of the maximum of each echo can be plotted versus the echo number. Some procedures using cross-correlation methods can also be implemented (Hull et al., 1985). In such approaches the major difficulty is to find a similarity criterion between successive echoes. Due to attenuation, high frequencies can be filtered out during propagation and the echoes become less and less similar during propagation. Applying the previous signal processing method becomes tricky. Furthermore, when echoes are overlapping, analysis in the temporal domain is totally impossible. In order to overcome such difficulties, working in frequency domains is suitable. Indeed, if the fast Fourier's transform of the echogram is performed, its modulus is a pseudo periodical curve constituted by several peaks. The distance between two peaks Δf is given in a first approximation by the following relationship (Rosenkrantz, 2007; Rosenkrantz et al., 2009; Sinha and Kaduchak, 2001):

$$\Delta f = \frac{C_L}{2d} \tag{1}$$

In this expression C_L represents the longitudinal ultrasonic velocity in the fluid and d is the pipe diameter. In order to evaluate C_L it is possible to plot the position of many peaks versus the peak indice. A linear adjustment directly gives C_L if "d" is known. But, as the modulus of the FFT is a pseudo periodical signal one can also perform a second fast Fourier's transform. This operation provides the time domain signal after filtration which is also constituted by several peaks. In this graph, the position of the first peak ($t_{\text{first peak}}$) is exactly the time of flight of the ultrasonic signal in the pipe. Hence, C_L is deduced from the simple relationship (2). Moreover, it can be demonstrated that this signal processing reduces the noise and remains applicable in case of overlapping echoes (Rosenkrantz, 2007; Rosenkrantz et al., 2009). Indeed, with this method it is possible to select in frequential domain the response essentially due to the fluid under study. A schematic representation of this signal processing is illustrated in Fig. 3.

$$C_L = \frac{2d}{t_{first\ peak}} \tag{2}$$

3. Results

3.1. Signal processing robustness

Two echograms were generated using the same celerity value of 1650 m s^{-1}. As we will see later, this corresponds to a coconut water with a SSC close to 32°Brix. The cell diameter was equal to 1 cm. In the first case, no noise was added and the exponential decay was small and comparable to a shear viscosity of 100 mPa s. In the second case, we chose a higher exponential decay (viscosity of 300 mPa s) and noise was added. Details concerning the link between viscosity and the exponential decay can be found in our previous study (Laux et al., 2014). These two echograms have been processed with the protocol described in paragraph 2.3.4.

The echograms and the time domain signal after filtration are shown in Fig. 4 with the value of velocity deduced. For coconut water, even when it is highly concentrated, the viscosity is less than 100 mPa s (see part 3.2.). Therefore, we can consider the error due to signal processing to be less than a few m.s^{-1} and constitutes a relative error of far less than one percent.

3.2. Coconut water

Typical echograms acquired on coconut water are presented in Fig. 5 for two different SSC. In Fig. 6 we have shown the viscosity measured with a rheometer and SSC in°Brix as a function of the ultrasonic velocity. Data obtained in (Laux et al., 2014) with the plane 25 MHz transducer in a plane configuration (accuracy $\pm$ 10 m.s^{-1}) and new results in the cylindrical geometry with the 5 MHz transducer are plotted together. For this new set of results the accuracy is also $\pm$10 m s^{-1}. This accuracy is composed of 3 terms: first, thirty measurements were performed independently after assembly/disassembly of the experimental bench. This leads to a standard deviation due to reproducibility of 1.5 m s^{-1}. Secondly one has to take into account the uncertainty on cell diameter. In our case it was 40 µm. Using classical formulas of errors propagation, the uncertainty reaches 4 m s^{-1}. At last, with the oscilloscope we acquired 10 000 points with a time base of 40 µs This introduces an error on time of flight measurement of 3 m s^{-1}. The quadratic sum of these various uncertainties leads to an error of $\pm$5 m s^{-1}. So, the accuracy for a confidence interval of 95% is equal to $\pm$10 m s^{-1}.

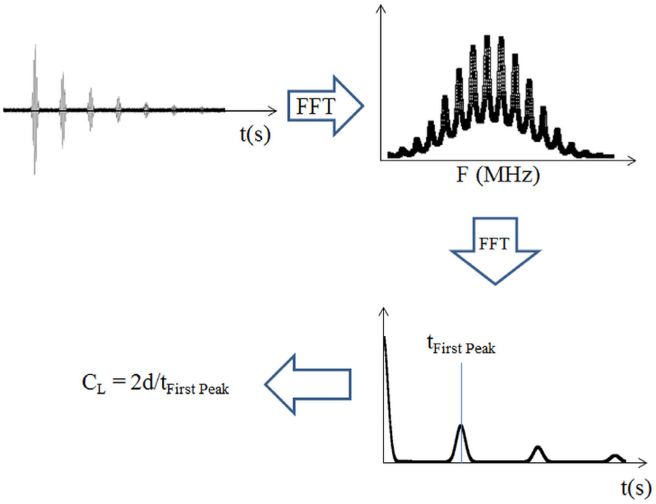

Fig. 3. Signal processing used for echograms analysis.

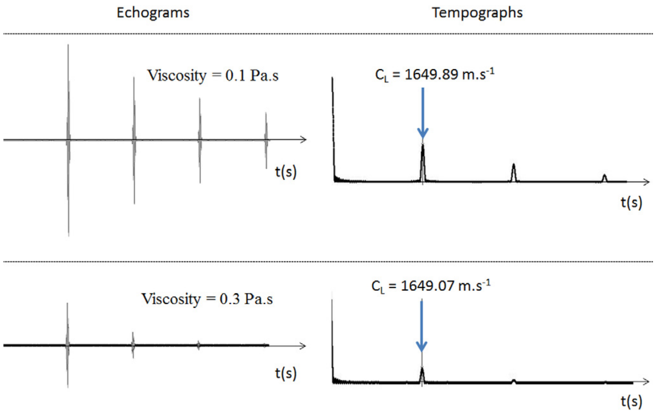

Fig. 4. Echograms simulated for signal processing testing.

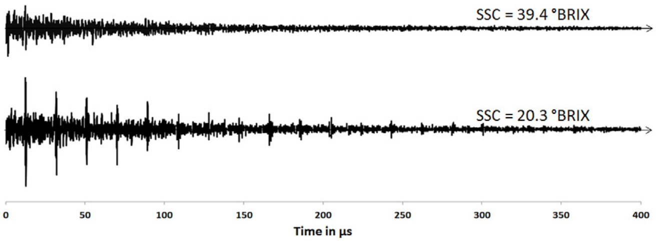

Fig. 5. Typical echograms acquired in coconut water for SSC = 20.3 and 39.4°BRIX.

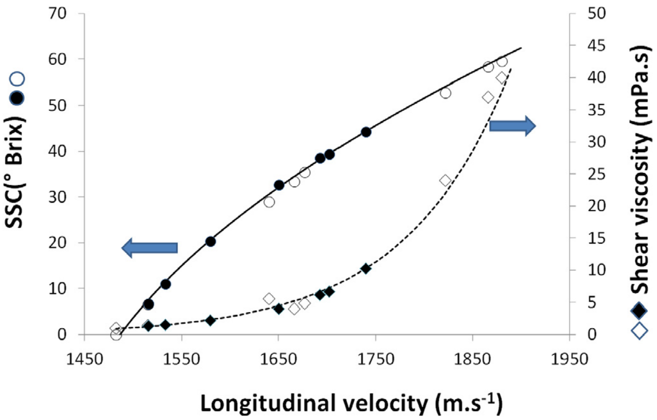

Fig. 6. SSC and Shear Viscosity of coconut water versus Longitudinal Velocity. Empty symbols: measurements obtained in 2014 at 25 MHz with a plane transducer immersed in the fluid. Filled symbols: present study at 5 MHz through a stainless steel pipe.

Hence, if the coconut water is concentrated, very small variations of SSC are detected. The trends observed in (Laux et al., 2014) are confirmed and we propose two non-linear adjustments. Regarding literature and especially the works of Contreras (Contreras et al., 1992), ultrasonic longitudinal velocity can be linked to SSC with a second order polynomial expression. In our case, if we take into account all data sets, the best polynomial adjustment ($R^2 = 0.99$) can be written as: $C_L = 0.0508(SSC)^2 + 3.5545(SSC) + 1486.8$. Consequently, after straightforward manipulations, SSC is linked to C_L with the relationship (3):

$$SSC(^{\circ}Brix) = -35 + \sqrt{0.2{*}C_L - 285} \tag{3}$$

The longitudinal velocity can be linked to the bulk modulus B and to the shear modulus G as follows: $\rho C_L^2 = B + \frac{4}{3}G,$ where ρ is the mass density (Zhao et al., 2003). If the liquid is purely viscous and Newtonian, and assuming that B is a constant, viscosity is a quadratic function of velocity (Zhao et al., 2003). Nevertheless such an adjustment for our data does not give good results for small viscosities. As our goal is the give working calibration laws, concerning the viscosity η in mPa.s versus ultrasonic velocity, an exponential trend has been chosen. It constitutes a very good adjustment ($R^2 = 0.99$) and is given by the relationship (4):

$$\eta = 10^{-6} e^{0.00928 C_L} \tag{4}$$

These two adjustments are shown in Fig. 6 in solid lines for SSC versus C_L and in dotted lines for η versus C_L.

4. Conclusion

Following a robust and simple experimental protocol for ultrasonic longitudinal velocity estimation, we have developed a simple manner to evaluate the shear viscosity and SSC (°Brix) of coconut water during its processing. For the two calibration laws established up to 60° Brix, the frequency used for the characterization does not appear to be a key parameter. Up to now, all characterizations have been carried out at 20 °C. In order to complete this calibration step, other relationships between ultrasonic velocity and temperature should be established. However, in an initial approximation, regarding the evolution of ultrasonic velocity versus temperature in water, sucrose/water mixtures, glucose/water mixtures … (Contreras et al., 1992), the order of magnitude will certainly be around 3 $m\ /(s.^{\circ}C)$. Furthermore, for sugar solutions it appears that the effect of temperature is quite the same for all sugar concentrations or SSC. Hence, using the relationship (3) it will certainly be possible to quantify the relative evolution of SSC during coconut concentration even if the temperature if not equal to 20 °C. These hypotheses will have to be confirmed using a new set of experiments.

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