Method for Acoustic Characterization of Materials in Temperature

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Abstract. Harsh environment applications like nuclear plant safety or oil and gas extraction need non-destructive testing methods to control the integrity of critical parts. Such inspections are commonly done by using ultrasonic Non Destructive Testing. However, it is difficult to design transducers for harsh environment due to the lack of reliable data about material properties. For instance, acoustic properties of frequently used thermoplastics and epoxies are poorly documented. In this paper, a method for characterizing materials between 40°C and 180°C is presented.

The characterization is carried out in immersion in a heated tank full of cooking oil. A transmission signal is obtained using two transducers, one transmitter and one receiver. This transmission signal is measured when a sample is present (perpendicular to the beam or not) or absent (reference case). Then, this differential method compares the two signals, and permits to determine acoustic velocity, impedance and attenuation. Using relations between mechanical properties and wave velocities, one can calculate mechanical properties like Young's modulus and Poisson's ratio. The originality of the method comes from the allowed temperature range (up to 180°C).

Two widely used materials have been fully characterized:

- Rexolite 1422, often used to make wedges at ambient temperature. This material is used as a validation for the method as the properties are quite well known at room temperature. It has been measured from 40°C to 100°C (Tg = 114°C), as shown below.

- Peek 1000, resistant to harsh environment such as high temperature and chemical environment. It has been measured from 40°C to 170°C (Tg = 150°C).

1. Introduction / State of the art

Design of transducers requires a complete knowledge of material properties [1-4]. For high temperature transducers, it is moreover necessary to know the variation of those properties with temperature. Those material properties are the speed and attenuation of both longitudinal and shear waves. Those values are necessary to simulate the behaviour of each of the transducer components in order to estimate the response of the whole transducer at different temperatures before fabricating and testing it.

The characterization of acoustic properties can be made by several methods [5-8]. One of them is ultrasonic method, which uses sound to characterize solid materials [9-13]. It can be used in transmission [10, 11, 14] or in pulse-echo mode [9, 13]. Usually, these measures are made at ambient temperature. However, some authors used this method to determine acoustic material properties at temperatures different from ambient [3, 4].

The originality of the method presented here lies in the fact that the acoustic characterization is made at different temperatures, from 40 to 180°C. In this paper, we used the ultrasonic transmission technique to characterize the acoustic properties of solid materials with temperature. The method presented here allows the determination of longitudinal and shear wave speed and attenuation in solid medium. Results obtained for Rexolite and PEEK will be presented.

2. Theory

2.1 Determination of sound velocity

Two ultrasonic transducers are immerged into a tank full of cooking oil, as shown in figure 1.

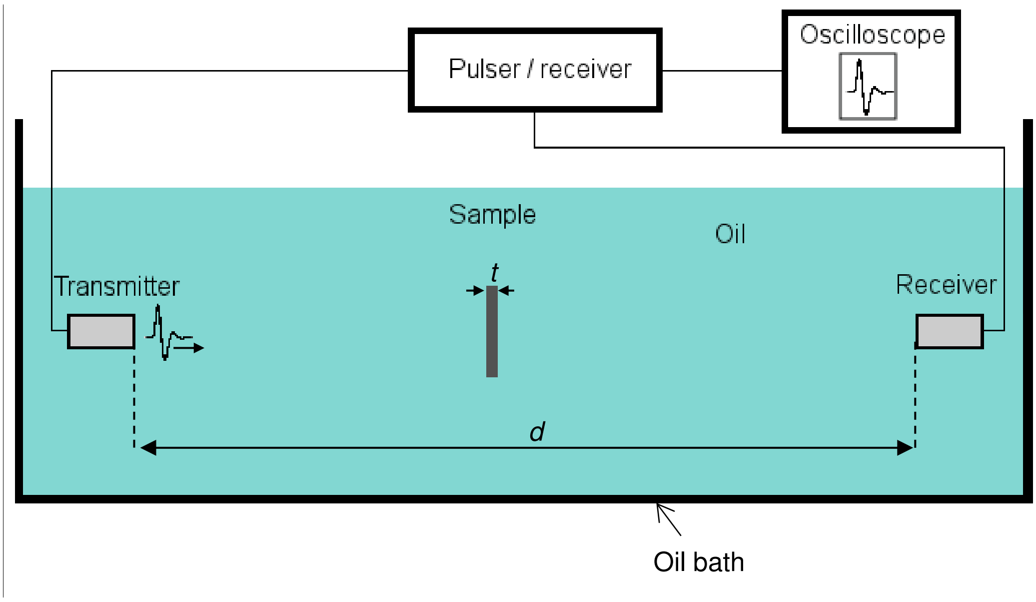

Figure 1 : Experimental setup. t is the thickness of the sample. d is the distance between the two transducers.

Characterization is done in two steps:

- First, the measure is made without the sample. This measurement is the reference case.
- Then the sample is added, and a new acquisition of the signal is made.

The determination of the difference of arrival times gives the speed of the longitudinal waves in the sample. The reference case gives the time of arrival for oil only T_{oil} in seconds, defined like the distance d in m between transducer divided by the speed of oil V_{oil} in m/s at a given temperature.

$$T_{oil} = {}^{d}/_{V_{oil}} \quad (1)$$

When the sample of thickness t in m is added, and the distance d is kept constant, the time of arrival for longitudinal waves T_{longi} in s is defined as:

$$T_{longi} = \frac{d-t}{V_{oil}} + \frac{t}{V_{longi}} \quad (2)$$

Then, by combining those two equations, longitudinal wave velocity V_{longi} in m/s is defined as:

$$V_{longi} = \frac{V_{oil}}{1 - \frac{V_{oil} \times \delta T}{t}} \tag{3}$$

Where δT is the difference of arrival times in s between the reference and the measure with sample. One could see that the speed of longitudinal waves in the sample is only determined by:

- The speed of ultrasound in oil,
- The difference of arrival times with a reference case, and
- The thickness of the sample.

This measure is independent of the distance, while it remains constant between the two measures.

When a plane sound wave in a liquid strikes a plane interface of a solid under oblique incidence, a reflected plane wave is generated, a longitudinal wave is refracted and a mode converted shear wave is generated into the solid by mode conversion [14]. If the angle of incidence is between the first and second critical angle, only shear waves are generated inside the sample.

To characterize shear wave speed, the same method as before is used but the sample is inclined as shown in figure 2, in order to produce only shear waves inside the sample.

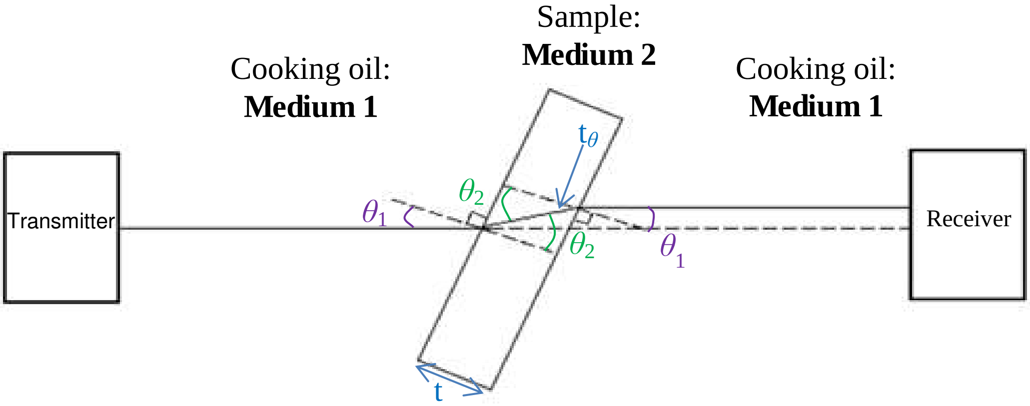

Figure 2 : Experimental setup for shear characterization

The speed of those shear waves V_{shear} in m/s is determined by (4), where θ_1 is the angle of incidence in radians, and t_θ is the distance travelled by the shear wave inside the sample in m. Due to the presence of the angle, this distance t_θ differs from t thickness of the sample.

$$V_{shear} = \frac{V_{oil}}{\sqrt{1 + \frac{V_{oil}\delta T}{t_\theta} \times \left(\frac{V_{oil}\delta T}{t_\theta} - 2 \times cos\,\theta_1\right)}} \tag{4}$$

Like previously, this measure is not dependent of the distance between the transducers. However, it stays dependent to the speed of oil.

2.2 Determination of ultrasonic wave attenuation

Using this method, it is also possible to determine the attenuation of ultrasonic waves in solid materials by comparing the amplitude of received signal with (A_{longi} in V) and without sample (A_{oil} in V). The longitudinal coefficient of attenuation α_{longi} in dB/m is defined as:

$$\alpha_{longi} = \alpha_{oil} + \frac{20}{t} \times log_{10}\left(\frac{T_{12} \times T_{21} \times A_{oil}}{A_{longi}}\right) \quad (5)$$

In normal incidence, at the first interface (medium 1 to medium 2), the transmission coefficient in amplitude T_{12} without unit is defined as:

$$T_{12} = \frac{2 \times Z_1}{Z_2 + Z_1} \quad (6)$$

Symmetrically, the transmission coefficient in amplitude at the second interface T_{21} (medium 2 to medium 1) is defined as:

$$T_{21} = \frac{2 \times Z_2}{Z_2 + Z_1} \quad (7)$$

There, one can see that it is necessary to determine the acoustic impedance Z in MRayl of the sample and of cooking oil according to the definition of acoustic impedance defined by (10):

$$Z = \rho \times V_{longi} \quad (8)$$

To determine acoustic impedance, it is necessary to determine the specific density ρ in kg/m^3 of sample and cooking oil. For the cooking oil, it has been measured using a pycnometer at different temperatures. For the sample, either the measure is made in laboratory if the shape of the sample allows it, or the values are extracted from literature. As the samples are solid, variation of specific density is relatively small with temperature, and will be considered constant in calculations to come.

The coefficient of attenuation for shear waves, α_{shear} in dB/m can also be determined by the same method, by inclining the sample. Moreover, the transmission coefficients are also dependent of this incidence angle. Taking this into account, one obtains formula (9). A_{shear} in the amplitude of received signal with sample inclined, in V and α_{oil} is the coefficient of attenuation in oil in dB/m.

$$\alpha_{shear} = \alpha_{oil} + \sqrt{1 - \left(\frac{V_{shear}}{V_{oil}} \times sin\,\theta_1\right)^2} \times \frac{20}{t_\theta} \times log_{10}\left(\frac{T_{12} \times T_{21} \times A_{oil}}{A_{shear}}\right) \quad (9)$$

Figure 2 shows that the beam is deviated by the inclined sample. The deviation D in m of the beam due to the passage into the inclined sample is determined by (10):

$$D = \frac{t \times \sin(\theta_1 - \theta_2)}{\cos(\theta_2)} \quad (10)$$

Considering a sample of thickness 1 cm, and a transversal velocity of 1200 m/s, with an incidence angle of 45 °, and a speed of waves in oil of 1000 m/s, D is equal to 1.91 mm.

The probe diameter is 10 mm, and the angle of divergence of the beam it emits (at 6 dB) is equal to 3 °. Considering the distance between transducers (usually 300 mm), the beam radius (at -6 dB) when it impacts the receiver is larger than 10 mm (calculus gives 15 mm).

In consequence, the deviation of the beam due to travel inside inclined sample can be neglected. However, it is important to note that this method gives only a maximum value of the shear wave attenuation coefficient, and not the exact one. For longitudinal attenuation coefficient, the coefficient is exact.

2.3 Oil calibration

The oil we used was cooking oil (hydrogenated coconut oil). It is commercialized under the name of vegetaline by Unilever Nederland. It is considered that this oil does not degrade with temperature cycles, so a calibration has been made. The speed and attenuation of longitudinal waves (no shear waves in liquid) in cooking oil have been measured precisely.

To measure the speed of longitudinal waves in cooking oil, the transducer's distance d has been divided by the time of travel. This measure has been made for several distances at each temperature, and the mean has been calculated. Those measures have been repeated for all temperatures. The results are presented in figure 3.

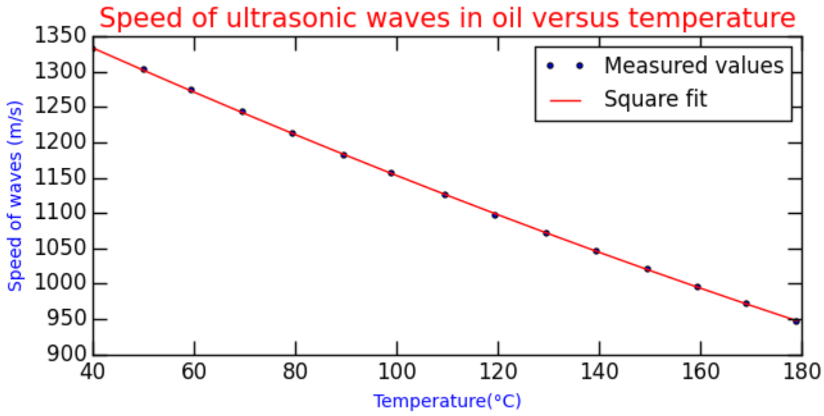

Figure 3 : Speed of ultrasonic waves versus temperature in cooking oil. The square fit gives Speed = 292.7E^{-3} * T² - 3.41895 * T + 1465.656

It is considered that amplitude decreases exponentially with distance. Working with acoustic decibels the attenuation of oil α_{oil} in dB/m can then be defined by formula (11):

$$\alpha_{oil} = -1 \times \frac{\Delta 20 \times \log_{10}(A_{oil})}{\Delta d} \tag{11}$$

A_{oil} is the amplitude of the signal in V at a distance d in m for a given temperature. The same calculus is made for each temperature and the curve of figure 4 is obtained.

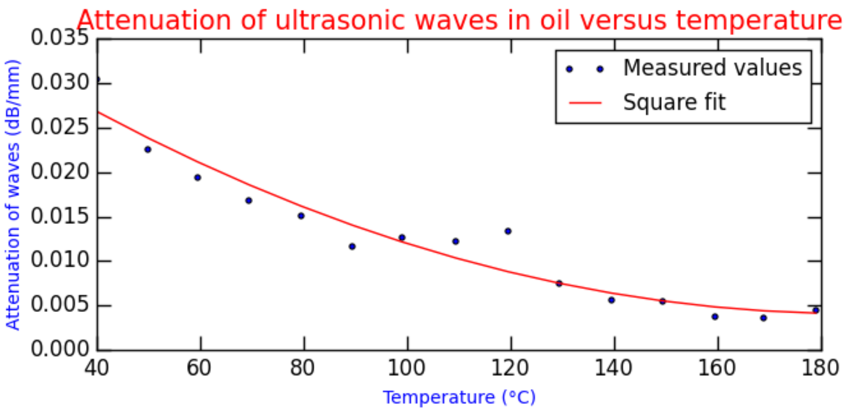

Figure 4 : Attenuation of ultrasonic waves (@2.3 MHz) in cooking oil versus temperature. The square fit gives Attenuation = 1.068E^{-6} * T² - 3.972E^{-4} *T + 4.102E^{-2}

The attenuation of cooking oil is quite low, but not negligible for low attenuation materials. It decreases with temperature. For those measures, it has been supposed that the ultrasonic beam shape modification due to distance and temperature could be neglected.

3. Experimental results and discussion

Two immersion-type homemade high temperature transducers with center frequency of 2.3 MHz have been used. The transmitting transducer was driven by a pulser (Panametrics 5900 PR) that produced short duration pulses of 8 µJ with a repetition rate of 200 Hz and 50 Ω electrical damping. The signal from the receiving transducer was sampled using an oscilloscope (Agilent technologies DSO5014A), which has an adjustable digital delay for triggering the sampling window. Each sampling window contained 1000 data points, and results of the average of 1024 measures. The data were saved in a text file and were treated by a python code. Oil bath temperature has been set stable during 1 h before all measures, to avoid temperature gradient in the tank (Memmert OB 45). Temperature of the bath was controlled by Pt100 temperature sensor (1 °C temperature set accuracy and ± 0.5 °C for temperature fluctuation). The temperature of oil was measured with a thermocouple type K.

3.1 Rexolite 1422

A cylinder of rexolite 1422 (cross linked polystyrene) from Ensinger UK with a diameter of 80 mm and thickness 10 mm was characterized. The specific density given by the supplier is 1.05 ± 0.01. Speed and attenuation have been measured and are presented in the following figures. It has been characterized from 40 to 100 °C to stay under the glass transition temperature.

Rexolite is a usual material for making wedges for contact NDT transducers. However, acoustic properties are just partially known. Supplier gives a longitudinal wave speed of 2350 m/s and a shear wave speed of 1150 m/s, at ambient temperature. By interpolating the data shown in figure 5, we obtained a longitudinal speed of 2340 m/s and a shear wave speed of 1140 m/s at ambient temperature. Taking into account errors of measurement, our measures are in good agreement with supplier’s one at ambient temperature.

Attenuation is not given by the supplier, but Wang and Cao measured it at 30 MHz and ambient temperature [2]. They obtained a longitudinal coefficient of attenuation of 1.1 dB/mm at 30 MHz. Considering that the variation of this coefficient with frequency is linear, it gives an attenuation coefficient of 0.084 dB/mm at 2.3 MHz. By interpolating the data shown in figure 5, we obtained an attenuation coefficient for longitudinal waves of approximately 0.15 dB/mm. The orders of magnitude are in good agreement. However, our value is slightly higher. This difference could come from:

- Material supplier
- Measurement method
- Calculus of transmission coefficients
- Wetting of oil

The behaviour of rexolite is constant with temperature from 40 to 100°C. However, it seems that above 90 °C, shear wave attenuation seems to slightly increase, which could be the beginning of the glass transition phenomenon (T_g = 113 °C).

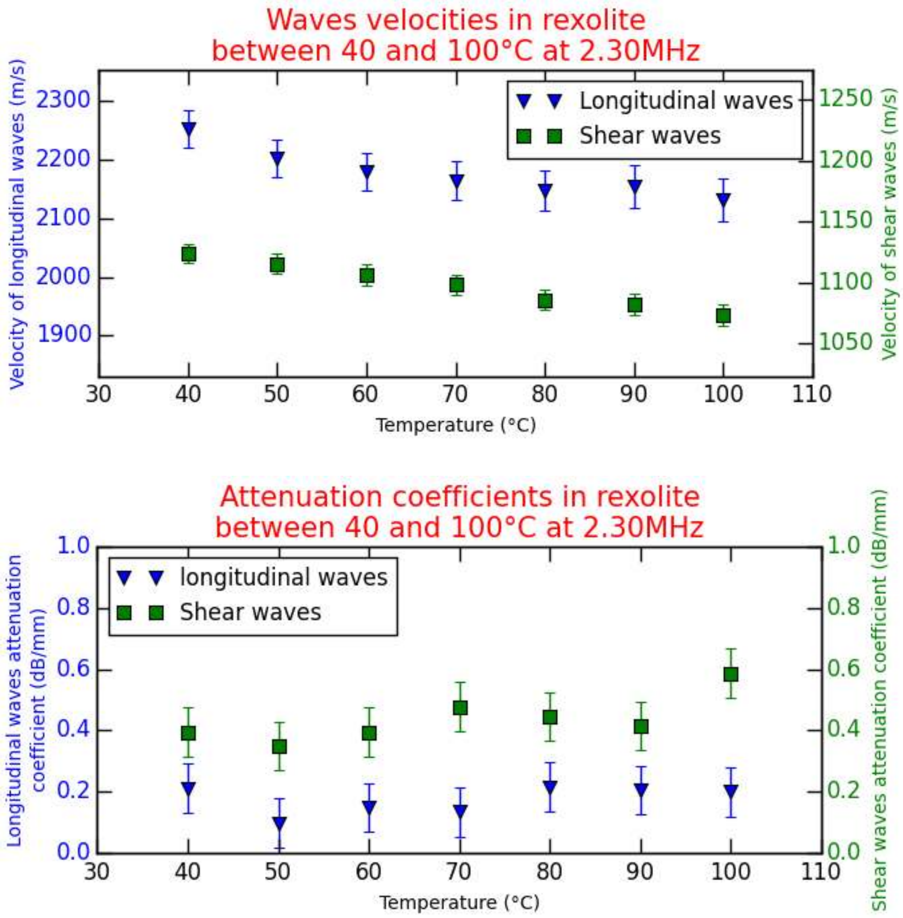

Figure 5 : Waves velocities and attenuation in rexolite between 40 and 100 °C

3.2 PEEK

A plate of ketron® 1000 PEEK (polyether ether ketone) from Quadrant Europe with dimension 150*100*8.65 mm (8.65 is the thickness). The specific density given by supplier is 1.31. Speed and attenuation have been measured and are presented in figure 6.

Carlson et al [4] obtained a longitudinal speed of 2555 m/s at 5 MHz and 37 °C. At 40°C, we obtained 2540m/s which is in good agreement (speed is constant with frequency). For attenuation also, the results are pretty consistent. Carlson gives an attenuation coefficient for longitudinal of 0.38 dB/mm at 37 °C and 5 MHz. Our measure gives 0.2 dB/mm at 40 °C and 2.3 MHz. At 5 MHz, considering the variation of attenuation coefficient as linear, our attenuation coefficient is approximately 0.4 dB/mm, which is in good agreement with Carlson's value, considering the errors. There is no measure at 180 °C due to a too high attenuation in tested material.

The behaviour of the material strongly changes around 150°C. This change is probably due to the glass transition phenomenon (glass transition temperature is estimated at 143°C according to the supplier). The speed decrease is stronger, and the attenuation increase above the glass transition, which is in good accordance with this phenomenon. Even if it has not been studied here, those values depend on the thermal story of the sample. Indeed, PEEK is a semi-crystalline material and its degree of crystallisation varies with thermal treatment, which impacts its mechanical properties

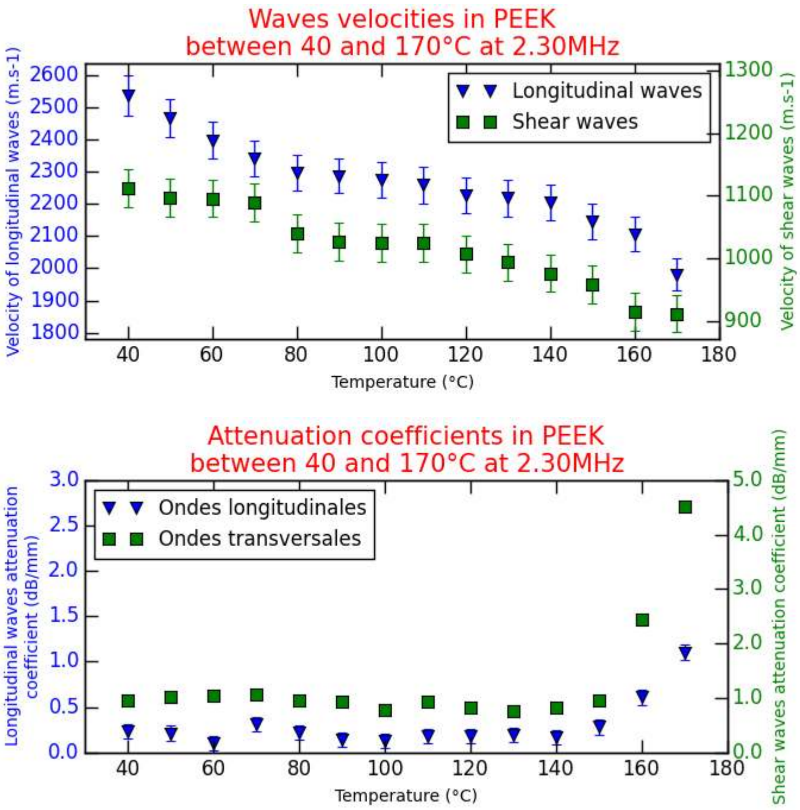

Figure 6 : Wave speed and attenuation in PEEK at 2.3 MHz between 40 and 170 °C

4. Conclusion

In this paper, we presented an improvement of the ultrasonic transmission technique, in order to characterize acoustical properties from 40 to 180 °C. This method presents several advantages. It is non-destructive and requires limited preparation of the sample. Ultrasonic velocity and attenuation can be obtained for both longitudinal and shear waves. As this method is relative, it is independent of the configuration if it remains unchanged during all measurements.

Rexolite and PEEK, two materials commonly used in ultrasonic transducer fabrication have been characterized. As we know, this is the first time those materials have been fully characterized at different temperatures.

The obtained values showed a good agreement with literature ones, measured at room temperature. Some differences with literature were observed for attenuation and some justifications were proposed.

References

[1] NANEKAR P.P., SHAH B.K. Characterization of material properties by ultrasonics. BARC Newsletter, Issue n°249.

[2] WANG H., CAO W. High frequency properties of passive materials for ultrasonic transducers. IEEE transactions on ultrasonics, ferroelectrics, and frequency control, 2001, Vol 48 n°1.

[3] MAK D.K, GAUTHIER J. Ultrasonic measurement of longitudinal and shear velocities of materials at elevated temperatures. Ultrasonics, 1993, Vol 31 n°4 pp 245-250.

[4] CARLSON J.E., VAN DEVENTER J., SCOLAN A., CARLANDER C. Frequency and temperature dependence of acoustic properties of polymers used in pulse echo systems. IEEE ultrasonics symposium, 2003, pp. 885-888.

[5] ASTM. Standard Pratice for Measuring Ultrasonic Velocity in Materials. E494-10 2010, 14p.

[6] BOUHADJERA A., SCHUBERT F. An ultrasonic mode conversion technique for characterizing prism-shaped material samples – experimental and numerical results. ECNDT, 2006, Berlin.
[7] AUDOIN B., ROUX J. An innovative application of the Hilbert transform to time delay estimation of overlapped ultrasonic echoes. Ultrasonics, 1996, Vol 34, pp 25-33.
[8] AGRAWAL M. et al. Characterization of mechanical properties of materials using ultrasound broadband spectroscopy. Ultrasonics, 2016, Vol 64, pp 186-195.
[9] HOSTEN B., BAUDOUIN S. Ultrasonic measurements of elastic constants in polymeric matrix/glass fibers composite materials versus temperature. Review of Progress in Quantitative Nondestructive Evaluation, 1995, Vol. 14, pp. 1209-1216.
[10] HE P. Direct measurement of ultrasonic dispersion using a broadband transmission technique. Ultrasonics, 1999, Vol 37, pp. 67-70.
[11] PETERS F., PETIT L. A broad band spectroscopy method for ultrasound wave velocity and attenuation measurement in dispersive media. Ultrasonics, 2003, Vol 41, pp 357-363.
[12] MASON W.P, McSKIMIN H.J et al. Physical acoustics, Principles and Methods. Vol 1A. USA: Academic Press Inc. Ltd.1964, pp 272-330.
[13] RODRIGO M.A.G, The ultrasonic pulse-echo immersion technique and attenuation coefficient of particulate composites [online]. Mechanical engineering and applied mechanics. University of Rhode Island, 2013, 141 p. Available on http://digitalcommons.uri.edu/dissertations/AAI1549142 (consulted 02/2016)
[14] GREENBERG Y., YAHEL E., GANOR M., et al. High precision measurements of the temperature dependence of the sound velocity in selected liquid metals. Journal of Non-Crystalline Solids, 2008, Vol 354, 4094-4100.
[15] BELGROUNE D., DE BELLEVAL J.F., DJELOUAH H. A theoretical study of ultrasonic wave transmission through a fluid-solid interface. Ultrasonics, 2008, vol 48, pp. 220-230.
[16] HUANG N.E., SHEN S.S.P., Hilbert-Huang transform and its applications, Vol5. Singapore : World scientific publishing Co. Pte. Ltd, 2005, 324 p.