Time evolution of ultrasonic parameters variations for a biphasic emulsion during the phases separation process

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Abstract. *This research studies the time evolution of acoustic propagation parameters of an emulsion during the separation process into its original phases. The main aspect of this work is to deal with the behavior of nonlinear, nonhomogeneous and time-varying media when they are submitted in an acoustic field. The study is based on spectral analysis theory of ultrasound backscattered echoes for understanding of physical process of the wave-emulsion interaction. The biphasic emulsion, which has been analyzed during this study is composed by a mineral oil (phase A) and vegetable oil (phase B). The methodology was based on time-frequency analysis of backscattered broadband ultrasound echo at center frequency of 5 MHz and bandwidth of 8 MHz @ -3dB using an ultrasound system in pulse-echo mode. The attenuation coefficients were calculated for each separated phase resulting in 0.82 dB/cm-MHz to phase A and 0.59 dB/cm-MHz to phase B. The B/A parameter have been evaluated resulting in 8.43 (phase A) and 10.74 (phase B). The acoustic parameters variation allowed identified different emulsification state inside emulsion. The time evolution for the emulsion was described through the increase of attenuation and B/A . The acoustic propagation velocity does not provide relevant information about the phases separation process, because it showed a minimal variation. The mean percentage variation for the time evolution of the acoustic parameters was 1.39 % to the acoustic propagation velocity, 46.9 % to* β *and 51 % to B/A. We hope that observations can be extrapolated aiming a better understanding of ultrasonic waves interaction in non-homogeneous time-varying medium.*

Keywords: *Acoustic Parameters,Biphasic Emulsion,Inverse Emulsification Process,Spectral Analysis,Time Evolution.*

1. INTRODUCTION

The technology and knowledge have expanded adequately the use ultrasound as one of the most important techniques applied for materials characterization with high performance and low cost. The materials characterization by ultrasound is currently a fruitful field in the control processes and materials analysis for many industries and medicine.

The ultrasonic inspection techniques studies systematically the ultrasound energy interaction with the material. Sometimes this inspection is non-quantitative but, frequently, these measures evaluate wave physical parameters from ultrasound echoes. Therefore this ultrasound inspection become a quantitative analysis Njeh *et al.*(1999), Pazin *et al.*(2003).

Parameters such as the acoustic propagation velocity and attenuation coefficient are the main base techniques for the evaluation material properties Dukhin *et al.*(2005). Also, several researches have focused on nonlinear measure characterization of an acoustic propagation which can help to understand the effect of wave distortion, formation higher harmonics and acoustic saturation that limit the energy transference for a material when it is submitted to a high intensity acoustic field Dong *et al.*(1999).

In this context, this work evaluate the time evolution of the acoustic parameters, both linear and nonlinear, from a biphasic oil emulsified using an ultrasound pulses of 5 MHz. This experiment used a medium with two immiscible liquids in the process in which they are not emulsified until formation of emulsion. The system configuration used for the capture of backscattered ultrasound signal is known as pulse-echo in which the transducer is coupled through water into acoustic tank containing the sample of biphasic emulsion. Thus, the estimative of acoustic parameters during the time evolution of the biphasic emulsion was based in techniques as described for Zhang *et al.*(1990), Junru *et al.*(1997) and kourtiche *et al.*(2001). This work used a comparative spectral analysis among many ultrasonic echoes from the biphasic emulsion interface formation and a reference medium (water). The variations of power spectral density for echoes from biphasic emulsion allowed the evaluation of attenuation and nonlinearity (B/A parameter). The originality of this work is to deal with acoustic parameters and the evaluation of emulsions in nonlinear regions when changes to its original phases. In this study it is intended to understand the acoustic wave interaction with the biphasic emulsion during its time variation.

2. ACOUSTIC PARAMETERS

The acoustic propagation velocity for a medium is evaluated as the rate of time delay through the material thickness. The used technique consists in emitting an ultrasound pulse and then measure the acoustic time of flight when the echo is received at transducer Povey (1997). The velocity $\boldsymbol{v}$ is determined by the expression:

$$\boldsymbol{v} = \frac{2\boldsymbol{d}}{\boldsymbol{t}} \tag{1}$$

where $\boldsymbol{d}$ is the material thickness and $\boldsymbol{t}$ is the time of flight. In homogeneous medium the linear model of acoustic propagation velocity is $\sqrt{B/\rho_0}$ where B is the *adiabatic bulk modulus* for a fluid, wikipedia (2007). For a mixture the acoustic propagation velocity depends essentially of the density and compressibility values Dukhin *et al.*(2005). This is important for the interpretation of the relation between the ultrasonic velocity and the stability phases that compose the emulsion.

2.1 Attenuation

The acoustic attenuation is the wave energy loss when it propagate throught a medium. The attenuation values in materials characterization can be used to differentiate one medium from another. For the attenuation evaluation, the emulsion is considered like a system that receives an input signal and produce an attenuated signal output. This situation can be seen by the following expression Eq.(2) in the frequency domain Soetanto *et al.*(1990):

$$Y(x,f) = A(x,f) * U(x,f) \tag{2}$$

where $Y(x,f)$ is the attenuated spectrum for the output signal (echo) and $U(x,f)$ is the spectrum for the input signal (acoustic excitation) in function of the material thickness x and the signal frequency f. $A(x,f)$ is the material transfer function and it can be assumed as:

$$A(x,f) = e^{\beta(x)f} \tag{3}$$

where β is the accumulative attenuation coefficient of the material thickness $\boldsymbol{x}$.

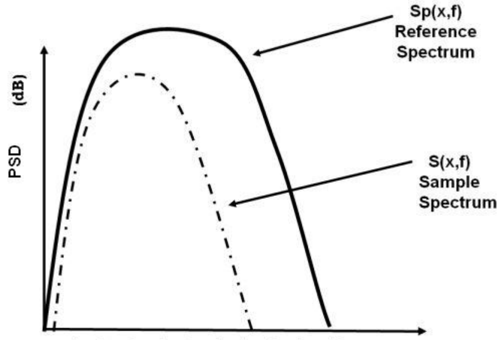


Figure 1. Low pass effect on the attenuated spectrum *S(x,f)*.

For the evaluation of attenuation where it is not necessary to assume that an ultrasound inspection pulse is gaussian is the comparative method. This consists in the division among the spectrum *S(x,f)* for an echo from emulsion interface formation and the spectrum *Sp(x,f)* for a specular reflection from medium reference see Fig.2, Treece *et al.*(2004). The Equation (4) illustrated this situation:

$$\frac{S(x,f)}{Sp(x,f)} = A(x,f) \tag{4}$$

The attenuation is given for spectral division and therefore for the spectrum *S(x,f)* can be seen a central frequency reduction when it is compared with the reference spectrum *Sp(x,f)*. This effect is seen in the Fig. 1 as a low pass effect for *S(x,f)*. The comparative method of spectral division produces an attenuation curve for the emulsion in the frequency domain. Thus, the evaluation of the attenuation coefficient is made applying a linear regression on the attenuation curve.

The attenuation coefficient value β is calculated based in the adjusted model for the attenuation transfer function that can be seen in the following expression:

$$ln(\frac{S(x,f)}{Sp(x,f)}) = -\beta f + b \tag{5}$$

where the term $-\beta f$ correspond to term *mx* in the straight line equation and the value parameter ***b*** corresponds to the value in the ordinate axis. According the previous, the acoustic attenuation can be seen as a power relation $\alpha = \beta f^n$(Shaffer *et al.,*1984), where α is the attenuation expressed in *dB/MHz* units, β is expressed in *dB/cm-MHz* units,***f*** is the frequency in (MHz) units and ***n*** is the value of frequency-dependent attenuation with ***n*** = 1.

2.2 Parameter of nonlinearity B/A

B/A is a parameter that measures the non linearity of the relation pressure-density for a medium and it is measured rewriting the equation of state for acoustic waves propagation in fluids as:

$$p = p_0 + A(\frac{\rho - \rho_0}{\rho_0}) + \frac{B}{2}(\frac{\rho - \rho_0}{\rho_0})^2 + ..., \tag{6}$$

where: p, p_0 are the instantaneous and the hydrostatic pressures, respectively; ρ is the density for the medium. A and B are terms of first and second order respectivily and they are related according to following expression:

$$\frac{B}{A} = 2\rho_0 c_0 (\frac{\partial c}{\partial p})_{o,s} \tag{7}$$

where c is the acoustic propagation velocity, the subscripts o and s denote the partial derivatives at equilibrium and at constant entropy, s, Brever(1959). The previous expression shows the dependence pressure of the medium stiffness.

The ratio B/A defines the medium capacity to distort a wave and to generate harmonics. Thus, the B/A magnitude for a medium will be increased if there are increases in the wave intensity, frequency and distance of propagation. In this study the B/A estimative was determined using the finite amplitude method. This method is based in the expression found for the second harmonic pressure amplitude p_2 of a finite amplitude wave propagated through a lossless fluid Bjørnø *et al.* (1986). This is defined by the following expression, Zhang *et al.*(1990):

$$p_2 = \left(2 + \frac{B}{A}\right) \frac{\pi f}{2\rho_0 c_0^3} x p_1^2(0) e^{-(\alpha_1 x + \alpha_2/2)x} \tag{8}$$

where x is the distance of propagation in the medium, f the fundamental frequency, $p_1(0)$ the pressure amplitude for the first harmonic to a distance $x = 0$, $p_2(x)$ the second harmonic value to a distance x and B/A is the parameter of second order of non linearity. The B/A value also can be measured with well precision employing a comparative method. This method compared the acoustic parameteres and properties between an sample medium and reference medium. The Equation (9) shows the expression used in this study to evaluate the acoustic parameter of nonlinearity B/A for the biphasic oil emulsified, kourtiche et al.(2001):

$$(B/A)_{sample} = 2 + \left(\frac{B}{A}\right)_{ref} \frac{x_{ref}}{x} \frac{\rho_0 c^3}{(\rho_0 c^3)_{ref}} \frac{p_2}{p_{2ref}} \frac{p_{1ref}^2}{p_{1x}^2} e^{\alpha_(1ref) - \alpha_(1)} \tag{9}$$

where $(B/A)_{sample}$ is the non linearity evaluated for the sample medium, $(B/A)_{ref}$ is the nonlinearity value of the reference, p_2 and p_{2ref} are the harmonic amplitude values obtained of the spectra for the sample medium and the medium reference respectively; p_{1ref} and p_{1x} are the amplitudes for the first harmonic in the reference and the sample medium respectively; x_{ref} and x are the thickness for the reference and the sample medium; $\rho_0 c_{ref}^3$ e $\rho_0 c^3$ are the respectives acoustic propagation velocity and density for the reference and the sample mediums.

In this study, the water was chosen as reference medium because it is an elastic medium that does not attenuate an ultrasound wave when it is propagated and also its acoustic propierties are known. Thus, calculate B/A require accurate, especially when the medium to study is nonhomogeneous because this lead a transmitted acoustic wave for a phase distortion which will may cause inaccurate measurement of attenuation and acoustic propagation velocity values producing consequently mistakes for the B/A measured.

3. MATERIALS AND METHODS

The system used for the acoustic parameters measurements is described for the following components: a) ultrasound equipment panametrics model 5800, b) ultrasound transducer panametrics of 5 MHz of 2.2 cm diameter, c) aluminum plane reflector of 5 cm thickness, d) acoustic tank with height of 18 cm e) support for the ultrasound transducer, f) biphasic oil Sample.

The Figure 2 shows the system used for the capture of ultrasound echoes in the reference medium (water) and the biphasic emulsion.

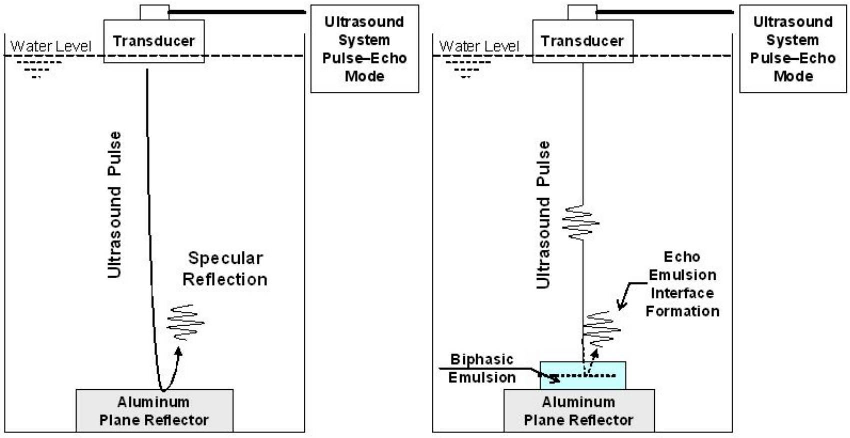
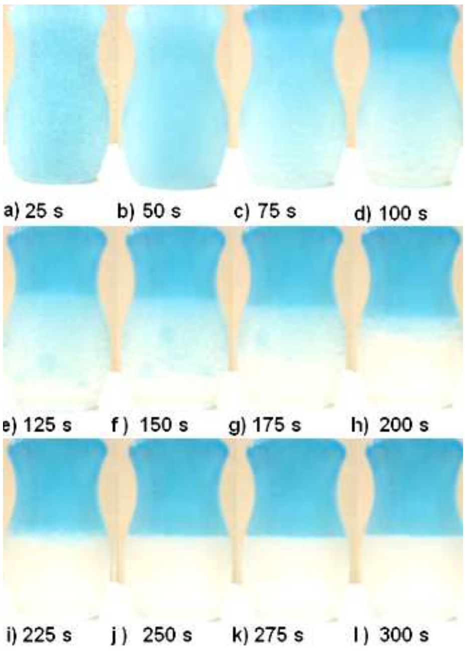


Figure 2. System diagram capture of ultrasound echoes.

Ultrasound echoes were recorded using an oscilloscope with a sampling rate of 500M/S. The specular reflection from an aluminum plane reflector was collected when the ultrasound pulse propagated through water column of height 18 cm. The specular reflection was considered as reference signal.

Later for the evaluation in the biphasic oil, it was emulsifed shaking it manually and then submitted to ultrasound field in pulse-echo mode during the phases separation process called in this study of inverse emulsification process. The time monitorization during the inverse emulsification process was of 300 seconds, so were collected twelve ultrasound signals from biphasic emulsion with intervals of 25 seconds. The Figure 3 illustred the inverse emulsification process for the biphasic oil during dynamic process of the stabilization into its original phases.


a) 25 s b) 50 s c) 75 s d) 100 s

e) 125 s f) 150 s g) 175 s h) 200 s

i) 225 s j) 250 s k) 275 s l) 300 s

Figure 3. Inverse emulsification process in the time

The power density spectral (PSD) was calculated for the specular reflection and for the twelve echoes recorded from emulsion interface formation. Theses were compared and analyzed using spectral analysis. The Figure 4 shows the spectra for the specular reflection and for an echo obtained from emulsion interface formation.

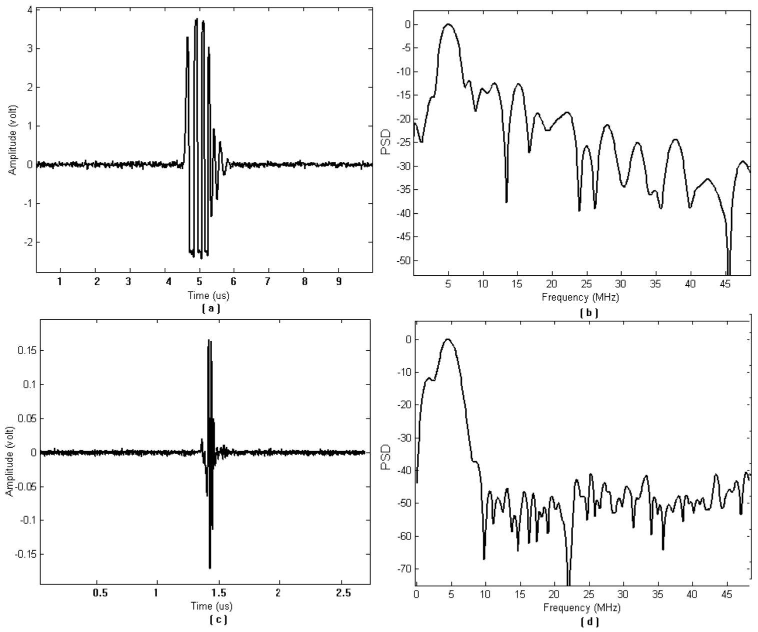


Figure 4. Specular Reflection and echo from interface emulsion with their spectrums respectively.

The PSD evaluation was made using the periodogram calculate from a subroutine in Matlab (Math Works, Inc.), so for each echo proceeding from emulsion interface formation, the PSD evaluation was estimated using a window blackmanHarris with a same size as ultrasound pulse width measured in the time domain. The comparative spectral analysis technique allowed to compare the evaluated spectrum for the specular reflection in the reference medium with the evaluated spectrum for each echo proceeding from emulsion interface formation during the inverse emulsification process. Thus, this technique based in the Eq.(4) was used to calculate the attenuation curves for the twelve echoes from emulsion interface formation during the inverse emulsification process. They were adjusted using a linear regression aiming the attenuation coefficient estimative.The attenuation coefficient value β is the slope value of the straight line divided by the biphasic emulsion thickness.

The parameter B/A was evaluated according to comparative values of the second harmonic amplitude seen in the spectra obtained for echoes from biphasic emulsion interface and the reference medium. Therefore B/A was evaluated substituting in the Eq.(8) the estimated values β, the acoustic propagation velocity and the density of the the biphasic emulsion and the reference medium. In this study also were calculated the acoustic parameters for the phases separated A and B. The density values for the two types of oil tested were: 0.845 g/cm3 (phase A) and 0.101 g/cm3 (phase B).

4. RESULTS

The result of the acoustic parameters obtained for the phases separated A and B are shown in the Tab.1:

Table 1. Values obtained of the acoustic parameter for the oil phases "A" and "B".

MATERIAL	α (dB/cm)	β(dB/cm-Mhz)	Velocity Propagation (m/s)	B/A
Oil A	4.1	0.82	1516	8.43
Oil B	2.95	0.59	1583	10.74

The acoustic propagation velocity average found for the oil phases called "A" and "B" were in a similar range to the acoustic propagation velocity for the water, estimated in 1512 m/s. The attenuation values found for these oil samples were determined based in the angular coefficient value from the attenuation curve. The first phase oil "A" had an attenuation 28.04 % greater that The second phase oil called "B". According to the Tab.1, the B/A values were next to values found in the literature for the soybean oil: 10.27 and 9.30 Kim *et al* (2002) which have similar density values to the biphasic emulsion analyzed in this study. The phase oil with low attenuation presented a B/A value greater than the opposite phase oil see Tab.1.

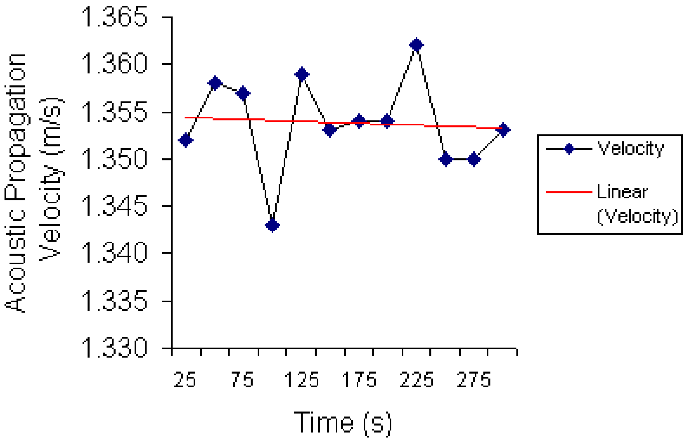


Figure 5. Time evolution acoustic propagation velocity

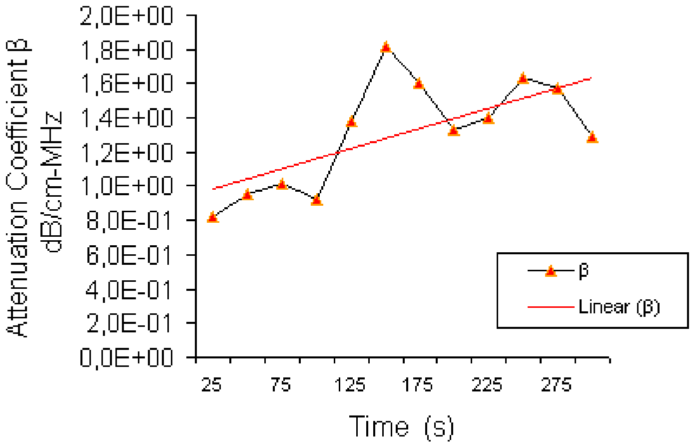


Figure 6. Time evolution Attenuation Coefficient

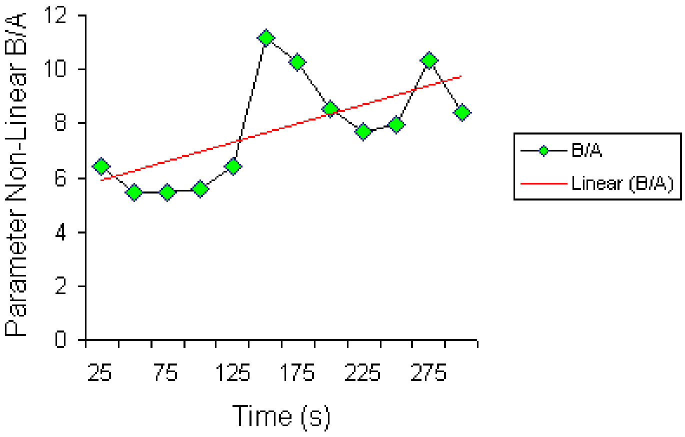


Figure 7. Time evolution B/A

The acoustic parameter results obtained for the biphasic oil emulsified are shown in the Fig.5.,Fig.6. and Fig.7. The profile of the acoustic propagation velocity during the separation phases process had a small variation and therefore the velocity was taken like constant in the time evolution study, aiming to reduce errors in the B/A mesured.

The time evolution of the attenuation coefficient β showed an increase according to the phase stabilizations of the biphasic emulsion. This probably is related with the break of the emulsion which produced an interface region given by the difference of impedances among the phases generating an echo. Thus, low attenuation values were observed during the initial process of emulsification because the energy dispersed for the oil bubbles size are smaller than the length wave of the ultrasound beam, therefore, the scattering multidirectional of low amplitude is not dramatic. In the final seconds of the phases separation process, was observed the increase of the attenuation values due the interface formation which is more attenuating tha the oil globules(phase disperse) seen in the first second of the emulsification process. The attenuation effect was seen for the spectra obtained from emulsion echoes during the time evolution. The effect low pass was exhibited when these spectral were compared with the reference spectrum. This effect was occasioned for the high frequency attenuation for the biphasic emulsion.

The parameter non-linear B/A showed values increasing to the long the time with a variation of 51 % between their minimum and maximum values. The time variations of these values were among a range reasonable according to values of non-linearity measured to different kind of oils, which were found in Kujawska et al. (2003) to oil linseed (similar oil vegetable) and oil soy Kim et al.(2002). The results showed during the first seconds that the increase of the attenuation reduces the shock formation and the distortion wave in the emulsion, therefore relatively a decrease of the B/A values were observed. So, the attenuation increase reduce the shock formation due to lost energy and for low attenaution values exist a tendency to high values of non-linearity. The previous is according to studies corroborated for Cartensen et al.(1986).

5. CONCLUSIONS

This study analyzed the interaction between the ultrasound waves and the biphasic emulsion composed by two immiscible oils during their separation process in the time. The relationship among the parameter variations: acoustic attenuation and B/A showed that they can be used to describe the phases separation process of the emulsion observing first that, when the medium have many oil bubbles, this can be attenuating because of energy loss, which is given by the dispersed oil in the emulsion. After, can be seen the existence of a nonlinear region with low values of non-linearity B/A that decrease with the increase attenuation during the first 125 seconds. When the biphasic emulsion exhibited nonlinear regions with low attenuation, the shock formation allow to evaluate quantitatively the medium through B/A estimative detecting amplitudes of the second harmonic. The acoustic propagation velocity variations not provide relevant information due to their small variations, which not allowed correlate the results with other parameters. The variations of β and B/A showed that the variations were among a range reasonable similar to acoustic parameter measurements of materials with oil composition. The monitoring of acoustic parameters during the time evolution provide information quantitative about emulsification state by the biphasic medium, so, the time evolution of the phases separation process was clearly identificable through the attenuation and B/A curves.

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