Characterization of a Time-Varying Medium by Ultrasonic Parameter Variations

Villamarín M. Julián A.
Department of Electrical Engineering
School of Engineering - University of São Paulo,
São Carlos, Brazil
julianvm@sc.usp.br

Mosquera S. Jaime A.
Department of Electrical Engineering
School of Engineering - University of São Paulo,
São Carlos, Brazil
jamosquera@usp.br

Abstract—Ultrasonic parameter variations were measured from a nonhomogeneous time-varying medium aiming to describe dynamic changes in the structure of the sample study by the relationship among ultrasonic parameters. The sample in study was an immiscible biphasic liquid mixture, which was emulsified and then monitored by ultrasound exposure in pulse - echo mode during time of phases separation. Quantitative ultrasound (QUS) techniques based in spectral analysis were employed to extract ultrasonic parameters from backscattering ultrasound echoes recorded in the interest region (ROI). The time-evolution of ultrasonic parameters estimated led to the identification of different changes in the ROI. These parameters let to understand about how together attenuation, the nonlinear parameter and IBC are involved with property changes in the ROI. The IBC was the best parameter to describe the behavior during the most emulsified state in the sample study. The attenuation coefficient β and B/A allow to identify the time of emulsion break-up.

Keywords-Emulsion, Backscattering, Nonlinear parameter, Spectral Analysis, Ultrasonic Parameter.

I. INTRODUCTION

Non-destructive ultrasound testing is used as a potential characterization tool in industrial processes [1, 2] and clinical applications such as quantitative characterization of biological tissues [3]. Quantitative ultrasound (QUS) techniques systematically study the interaction of ultrasound energy with the material in question through the extraction of ultrasonic parameters. Thereby, it is possible to provide information about the structural heterogeneity, composition and physical state of analyzed samples. QUS techniques can detect the presence of inhomogeneities in liquid mixtures as well as predicted structure changes in complex media [4]. Conventional ultrasonic parameters such as velocity or attenuation are commonly used in ultrasound characterization techniques because these parameters are very sensitive to the differences in physical properties of the liquid components [5]. Ultrasonic attenuation serves to differentiate one medium from another, to determine alterations in materials, to characterize dispersion in colloid systems and it is useful in emulsions characterization. In addition, ultrasonic parameters as for example, the integrated coefficient of backscattering (IBC) and the nonlinear parameter B/A are also commonly estimated to provide additional information of media with complex chemical composition. Currently, the ultrasonic parameters continue to be measured at rest and dynamic media using theoretical models about different scattering propagation process which are very discussed for several authors [6]. Most of studies concerning QUS characterization techniques aimed their research in materials at rest to measure acoustic parameter variations in substances, biological tissues and structures with particle aggregates such as emulsions [7]. Otherwise, ultrasonic techniques began to be widely used in dynamic media to improve industrial processes such as, the monitoring processes of polymerization and crystallization in liquid mixtures. Thereby, innumerable studies have reported success involving ultrasonic measurements in time varying media such as, the monitoring of the dynamics of condensing and non-condensing of liquid films by time-of-flight measurements [8], real-time assessments of membrane fouling during liquid separation processes [9], and monitoring of ultrasonic wave propagation in crystallizing mixtures during industrial production of pharmaceuticals and agrochemicals [10]. Therefore, ultrasonic parameters provide a better in-sight in the monitoring of physical state and characterization of dynamical phenomena for complex media, liquid suspensions - colloids and emulsions.

This study aims to evaluate ultrasonic parameter variations in a time-varying liquid mixture attempting to obtain knowledge about the relationship among ultrasonic parameter variations and how they will shed light to describe dynamical changes in the medium structure. Ultrasound backscattering echoes were collected from biphasic liquid in study. The echoes were analyzed by QUS techniques based on spectral analysis. Thus, the following ultrasonic parameters were measured: speed of sound (SOS), frequency-dependent attenuation coefficient, nonlinear parameter B/A and backscattering coefficient (IBC). The experimental results fell within a reasonable range of acoustic parameter measurements that were reported in this area of study [11].The quantitative analysis enabled us to identify the phase stabilization process in the liquid mixture inasmuch as parameter variations described dramatic changes in the interest region (ROI) such as, for example, the time emulsion break-up. We expect that analysis of the results in this study will shed light to tackle characterization of dynamic media by relationship among ultrasonic parameters variations.

II. MATERIALS AND METHODS

A. Experimental setup

The experimental setup consisted of: 1) Ultrasound system *Panametrics* model 5800 with output energy of 100uJ; 2) Pulser / receiver transducer with a center frequency of 5 MHz and a bandwidth of 2 MHz @ @3dB; 3) *Tektronix* TDS 3034B 300 oscilloscope to record ultrasound signal at a sampling rate of 100M/S.

The experimental sample was a biphasic liquid composed of: 1) Mixture of mineral and vegetable oils (Phase A) and 2) Mixture of propylene glycol with water (Phase B). The density and volume estimated to phase A was 0.840g/cm3 and 67 ml respectively, while that of phase B were estimated at 1.010g/cm3 and 60 ml.

Initially, the algorithms implemented for the quantitative ultrasound analysis were calibrated for ultrasound propagation in water, considered as reference medium. The ultrasonic parameters such as β and B/A were determined for A and B phases by the substitution method [12] at room temperature (25ºC). After A and B phases were mixed by mechanical agitation process. This procedure resulted in an unstable emulsion (after here this media will be reference as biphasic emulsion) whose phases began to separate over time. Biphasic emulsion was then placed inside an acoustic tank containing water and subjected to focused ultrasound beam during the time phases separation of 300 s approximately. Ultrasound backscattering echoes were recorded from the region where the interface is formed in sample study (ROI). Thereby, ultrasonic parameters for biphasic emulsion were determined by substitution method such as seen in Figure 1.

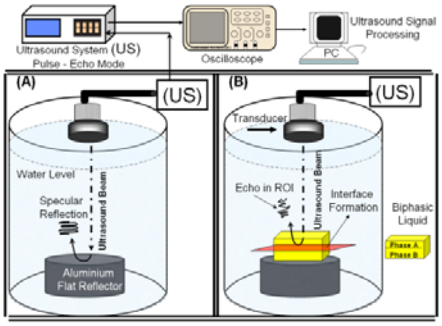

Figure 1. Substitution method. (A) Specular reflection (considered as reference signal) obtained from a flat aluminum reflector during wave propagation in water.(B) Ultrasound echo captured in the interface formation (considered as ROI) of the biphasic liquid.

Figure 2 shows the time evolution echoes for the biphasic emulsion during the separation process in their original phases. Also, in Figure 2 is possible to appreciate the emulsion break-up caused by droplets coalescence to produce a specular echo in the ROI.

B. Data Analysis Technique

The ultrasonic parameters β, B/A and IBC were estimated by a comparative spectral technique.

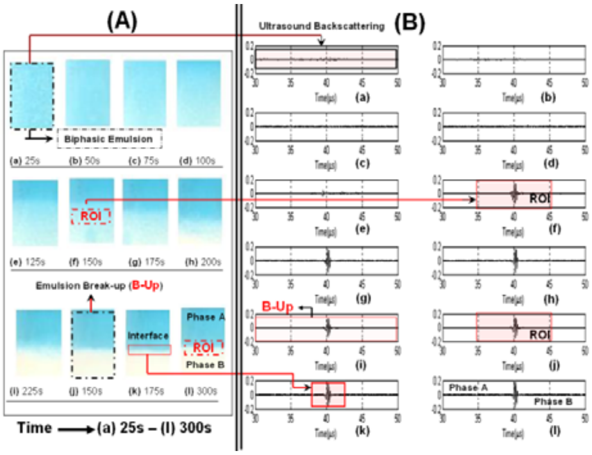

Figure 2. Time-evolution for biphasic emulsion. (A) Dynamical phases separating overtime (300s). (B) Ultrasound echoes collected in the ROI during phases separation over time.

This technique consisted of analyzing the power spectral density (PSD) between ultrasound echoes recorded from ROI - biphasic emulsion and the reference medium (water). The PSD was estimated from the periodogram using a Blackman-Harris window. Computational procedures for analysis QUS were developed using *Matlab* version7.6. The speed of sound (SOS) parameter was determined using the conventional time-of-flight (TOF) technique.

C. Estimation of Ultrasonic Parameters

TOF technique is commonly used in ultrasonic characterization of acoustic propagation velocity in fluids, food emulsion [1] and dairy products [2]. This involves the measurement of the time elapsed t for the wave to pass through the material's thickness. The mathematical expression is:

$$SOS = \frac{2x}{t} \tag{1}$$

where x is the thickness of the container holding the biphasic liquid; t is the time of flight for ultrasonic wave. Ultrasound attenuation was estimated to quantify the loss of wave energy when it propagated through the biphasic emulsion. The attenuation model expressed for an ultrasound plane wave through medium propagation is given by Equation (2):

$$|A(x,f)| = e^{\propto x} = e^{\beta f^{n} x} \tag{2}$$

where $A(x,f)$ is the module of amplitude attenuation transfer function for thesample; the power relation $\alpha = \beta f^{n}$ is the frequency - dependent term of attenuation coefficient; f is the signal frequency; $\beta$is the ultrasound attenuation coefficient expressed in dB/cm-MHz units; n is the value of attenuation frequency-dependence. In this study $n = 1$ due to the attenuation curveholds a linear relationship on the bandwidth of the transducer such as seen experimentally. The ultrasound attenuation was calculated by a spectral comparative method, which correct errors in the attenuation measurements caused by the effects of radiation coupling, signal pathway, and beam focus. In addition, it is not necessary to assume that an ultrasound pulse is Gaussian. The comparative method provides a standard measurement independent of the particular characteristics of the ultrasound device, such as the shape of the ultrasound wave. The comparative method consists of dividing a spectrum S by a reference spectrum S_{ref} estimated from a non-attenuated medium (water), see Fig.1. That is described by Equation (3):

$$\frac{S(x,f)}{S_{ref}(x,f)} = A(x,f) \qquad (3)$$

The attenuation coefficient for the sample study is calculated by applying a linear regression to spectral difference in the bandwidth of the transducer. It is depicted by Eq. (4) using Eq. (3):

$$ln\left(\frac{S(x,f)}{S_{ref}(x,f)}\right) = -\beta f + b \qquad (4)$$

For water, the attenuation coefficient is proportional to the square of the frequency. However, the attenuation value β is very small, around 0.0022 dB/cm-MHz and can be neglected. The nonlinear parameter B/A describes non-linear effects such as harmonic formation due to wave discontinuities in the time. This parameter allows to know how big is the distortion wave due to dynamical changes on biphasic emulsion, based in the pressure-density relation. The non-linear parameter was estimated by the comparative finite amplitude method [12]. This method compared known acoustic properties (density) and parameters between the biphasic emulsion and the reference medium. The nonlinear parameter B/A in the biphasic emulsion was determinate by Eq. (5):

$$(B/A)_{emul} = \left(2 + \left(\frac{B}{A}\right)_{ref}\right) \frac{\rho_o c_o^3}{(\rho_o c_o^3)_{ref}} \frac{p_2}{p_{2ref}} \frac{x_{ref}}{x} e^{-2(\alpha_{ref}-\alpha)} \qquad (5)$$

where ρ_o is the medium density and c_o is ultrasound velocity; p_2 is the second harmonic pressure. The subscripts ref and $emul$ refer to the reference and biphasic emulsion respectively. To obtain a quantitative evaluation of ultrasonic scattering in the sample study, the IBC was estimated. This parameter is defined as the mean backscatter power in a frequency range. IBC was calculated from the differential scattering cross-section per unit volume, called Backscattering Coefficient (BSC). The technique employed for this measurement compares the average PSD of backscattered ultrasound echoes (from ROI) and the PSD of a specular echo obtained from a planar aluminum reflector. This technique corrects the ultrasonic beam transmitted diffraction losses and cross-sectional area of the scattering volume. According to [13] BSC and IBC parameters can be estimated by Eq. (6) and Eq. (7):

$$BSC = \sigma(f,x) = \frac{1.45R^2}{A_0\Delta_z} w(f,x) \qquad (6)$$

$$IBC = \int_{f_{min}}^{f_{max}} \frac{\sigma(f,x)}{f_{max}-f_{min}} \partial f \qquad (7)$$

where R is the focal length; A_0 and Δ_z are the active surface transducerand the axial length of the gated volume respectively; $w(f,x)$ is the averagepower spectral density of the backscattered signal divided by the PSD of the reference medium.

III. RESULTS

Computational procedures to measure ultrasonic parameters were validated in water. The SOS measured in water agrees with literature and fell within a range of velocity values allowed for water (1492 m/s -1536 m/s) [11]. Table I shows experimental results for A and B phases and the reference medium. Acoustic parameters for the liquid mixtures: Phases A and B and reference medium.

TABLE I
ACOUSTIC PARAMETERS FOR THE LIQUID MIXTURES: PHASES A AND B AND REFERENCE MEDIUM.

Sample	**Ultrasonic Parameters**		
	***SOS* (m/s)**	***β* (dB/cm-MHz)**	**B/A**
Phase A	1516	0.82	8.43
Phase B	1583	0.59	10.70
Water (measured)	1512	0.0048	5.49
Water (Reported)$^{a-b}$	1520	0.0022	5.20

a. (ONDA CORPORATION, 2006), b. [11]

The results are depicted in Figures 3. (A), (B), (C) and (D) show ultrasonic parameters in ROI varying in time for SOS, β, B/A and IBC, respectively.

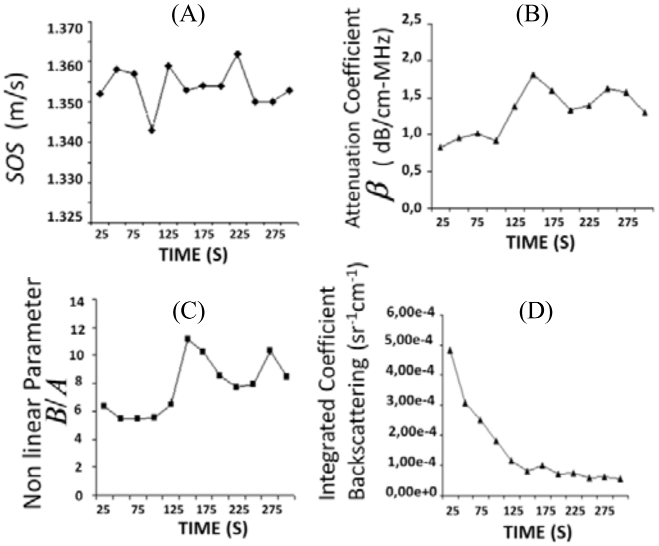

Figure 3. Time evolution of ultrasonic parameters. (A) acoustic propagation velocity SOS. (B) attenuation coefficient β. (C) nonlinear parameter B/A. (D) IBC.

IV. DISCUSSION

The SOS for biphasic emulsion showed minimal variations over time. The difference between the maximum and minimal variation of SOS was 1.4%. Hence, SOS variations according to dynamical changes in ROI were negligible. For calculations of the attenuation coefficient β was possible to assume a linear frequency-dependence of attenuation because the attenuation curve obtained by spectral difference holds to linear relationship on the band width of the transducer. Thereby, the law relation that described the frequency-dependence of attenuation for the biphasic emulsion was not necessarily a perfect model as oil-water systems and therefore the approximation linear was acceptable. The estimated attenuation revealed a low pass filter effect on biphasic emulsion spectra. This was because echogenic characteristic in the surrounding is not equal as the beginning. The dynamic scattering process from droplets coalescence and acoustic impedance changes in the time of the emulsion break-up (see Fig. 2) contribute to spectra attenuation. β values were fluctuating between 0.82 to 1.82 dB/cm-MHz and the highest variation registered to β was 46.9%.The B/A measurements in this study were made by the amplitude finite method using a fixed low energy of 100 uJ. During the first 125 seconds of the phase separation process, low amplitude waveform distortion were produced for oil droplets dispersion, which influenced the nonlinear propagation at sample study. However, time-evolution for B/A showed minimal changes before the time of emulsion break-up as seen in Fig. 3.(C). B/A values were shown to be larger during the time of break-up emulsion rather than beforehand. It is due to the formation of a more rigid structure in the ROI, which produces a distorted specular high-amplitude echo. Thus, the maximum variation during time-evolution of the nonlinear parameter was 51%.

The relationship between ultrasonic attenuation and the nonlinear parameter showed that an increase in attenuation reduces the wave shock form and influences the amplitude of component harmonics. Attenuation and B/A curves in Figures 3 (B) and (C) showed during the first 125 s opposite behavior. This difference is subtle due to the low echogenic characteristic in the ROI, which hold a relationship with low backscattering amplitudes. Therefore, high attenuation values tend to decrease slightly nonlinear B/A values. The time-evolution for IBC presented an expected behavior. The IBC decrease over time indicated the progressive disappearance of droplets in the biphasic emulsion. The estimated values were in the range of 0.48 x 10^{-5}cm^{-1} sr^{-1}and 5.64 x 10^{-5}cm^{-1}sr^{-1}.The time to break-up and subsequent phases stabilization can be seen in Fig. 3.(D), after 150 s where IBC values tend to remain constant. Also, IBC let to identify that the most emulsified state during the first instants before the time emulsion break-up. This is indicated by high IBC value due to highly scattered wave signals from droplets of the sample.

V. CONCLUSIONS

This study analyzed the interaction between ultrasound waves and a biphasic emulsion separating their phases in time. Ultrasonic parameter variations over time were obtained from the QUS processing of backscattering echoes in the ROI. The aim was to understand the relationship among ultrasonic parameter variations to explain the dynamic behavior of the medium. According to observed changes over time in the ROI, the time of emulsion break-up produced high variations for β and B/A in contrast with IBC, which shows low values. This is in agreement with the dynamic behavior of the sample because the impedance changes occurred in the time of break-up indicated that ROI-properties are not equal like the beginning. Therefore abrupt changes in the structure of the biphasic emulsion are represented by significant variations in the parameters. The relationship between the variations of the attenuation coefficient and B/A during most emulsified state showed a subtle difference among themselves. This was due to increased attenuation which decreases the harmonics propagation. Otherwise, the SOS parameter did not provide relevant information due to their minor variations, which prevented their correlation with the results of other parameters. The time-evolution for SOS was not significantly influenced by droplets dispersion. With exception to SOS, the relationship between the ultrasonic parameters allowed to describe different stages in the time-varying medium. The IBC was the best parameter to describe the behavior during the most emulsified stage and the time of phase separation. The attenuation coefficient and B/A allow to identify better than IBC the time of emulsion break-up. We extrapolated this study to analysis and classification of particles traveling in liquids by QUS techniques.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the Brazilian agencies CAPES (Coordinating Office for the Betterment of University Graduates. The authors also are grateful to Carlos Dias Maciel by his collaboration and suggestions to improve this work.

REFERENCES

[1] D. J. McClements, “Advances in the application of ultrasound in food”,analysis and processing, Trends in Food Science and Technology, vol. 61, pp. 293–299, 1995.

[2] A. S. Dukhin, P. J. Goetz, B. Travers, “Use of ultrasound for characterizing dairy products, Journal of Dairy Science”, vol. 88, pp. 1320–1334, 2005.

[3] J. Bellapart, J. F. Fraser, “Review: Transcranial doppler assessment ofcerebral autoregulation”, Ultrasound in Medicine & Biology, vol. 6, pp. 883–893,2009.

[4] T. Gomez, A. Arenas, L. E. Segura, E. de Sarabia, “Characterization of suspensions of particles in water by an ultrasonic resonantcell”, Ultrasonics , pp. 715–727, 2002.

[5] P. Combette, D. Roudil, G.Despaux, “Emulsion characterization by focused ultrasonic waves”, Ultrasonics, vol. 5, 329–334, 2001.

[6] H. Kim, T. Varghese, “Hybrid spectral domain method for attenuation slope estimation”, Ultrasound in Medicine and Biology, vol. 11, pp. 1808–1819, 2008.

[7] M. Hibberd, D., R. Challisb, “Ultrasonic monitoringof oil-in-water emulsions undergoing depletion occulation”, Journal ofColloid and Interface Science, pp. 77–87, 1997.

[8] Z. Chena, J. Hermansona, M. Shearb, P. Pedersenb, “Flow measurementand instrumentation”, Journal of Engineered Fibers and Fabrics,pp. 353–364, 2005.

[9] E. Kujundzic, K. Cobry, A. R. Greenberg, M. Hernandez, “Use of ultrasonic sensors for characterization of membrane fouling and cleaning”,Journal of Engineered Fibers and Fabrics, pp.35 – 48, 2008.

[10] T. Marshall, R. Challis, J. Tebbutt, “Monitoring and modeling of ultrasonic wave propagation in crystallizing”, Review of Quantitative Nondestructive Evaluation, pp. 1735–1741, 2002.

[11] M. C. Ziskin, P. A. Lewin, “Ultrasonic Exposimetry”, Crc Press,Inc.,U.S.A, 1993.

[12] D. Koutiche, L. Allies, A. Chitnalah, M. Nadi, “Harmonic propagation of finite amplitude sound beams: comparative method in pulse echomeasurement of nonlinear b/a parameter”, Meas. Sci. Technol, vol. 12, pp.1990–1995, 2001.

[13] M. F. Insana, R. F. Wagner, D. G. Brown, T. J. Hall, “ Describing smallscale structure in random media using pulse - echo ultrasound”, J. Acoust.Soc. Am. Vol. 6, pp. 179–192, 1990.