

Ultrasonic Attenuation Measurement Method for Characterization of Clay Particles Mass Fraction in Dilute Suspension.

A. Hamine^{1*}, B.Faiz¹, A.Moudden^{1,2}, A. Menou³,
A. Benallou³, D. Izbaim¹, G.Maze⁴ and D.Decultot⁴

¹ *Laboratoire Métrologie et Traitement de l'Information Faculté des Sciences, Agadir, Morocco*

² *Ecole Supérieur de Technologie Agadir, Morocco*

³ *ONDA/AIMAC, Aéroport Mohammed V, Casablanca, Morocco*

⁴ *Laboratoire Ondes et Milieux Complexe, Université du Havre, France*

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ABSTRACT

In the present work we have developed a technique based on the ultrasound for the quality control of barrier water, by measuring attenuation of ultrasonic waves in water slurries with various quantities of clay. This technique makes it possible to control in real time the presence of the clay grains whose dimension is about 10 μm , using an interface conceived under LabVIEW program. For slurries with different Weight percent of clay 1% or less, high sensitivity is gained by analyzing attenuation measurements obtained from multiple paths through the slurry. For slurries with higher concentrations of clay, sufficient sensitivity is obtained by analyzing data from a simple transmission. The experimental results show that the attenuation of sound due to particles varies linearly with mass fraction, and that the proposed theoretical model can be used to predict this attenuation.

Keywords: Clay density measurement, Ultrasonic On-line sensor, process control, Slurry concentration measurement, Weight percent solids measurement, Suspension characterization, Attenuation measurement.

* Corresponding author: A. Hamine

1. INTRODUCTION

Using ultrasonic measurements have been performed to characterize liquids, solids, and gases for over 52 years. Most works applies to ultrasonic process measurement, focusing on application for density, flow measurement, and interface sensing. The need for compact, non invasive, real time measurement of density and concentration for waste slurries of clay of barrier water fostered the development of an ultrasonic sensing technique based on the reflection and transmission of ultrasonic signal at the sensor-fluid interface. While commercially available ultrasonic attenuation spectrometers have been fully used. One problematic field of application is the size region for particles or droplets larger than 10 μm . another area of concern exists for spherically shaped or inhomogeneous particles. The stopping water contains various sizes of clay grains. The knowledge of the quantity of clay present in water will make possible to optimize the addition of the chemicals products which allow the clay recovery. The physical treatments of water is done in two phases, a preliminary phase is directed towards a natural elimination of the suspended matter by decantation and a secondary phase which is used for elimination of colloids and clays by addition of chemicals products, these two phases are controlled in

real time by this ultrasonic technique. The measurement of the viscoelastic parameters of water will enable us to work out abacuses, which will be useful for the quality control of water of stopping, and to optimize the addition of the chemical products necessary to recover clay particles.

The distribution of the particles and their size in slurry are connected, with the determination of the attenuation coefficient /1/, or/and the velocity of sound. Ultrasonic velocity is the distance which the wave cast through the sample for a unit of time. The attenuation coefficient is the measurement of the reduction in the amplitude of ultrasonic wave for a unit distance /2, 3/.

Ultrasonic velocity and the attenuation coefficient of the sample are given with same preceding manner, except we must replace the distance d by $2d$, because the wave made two alleys and return in the container /4/.

Ultrasonic velocity: $c = \frac{2d}{\Delta t}$, and the attenuation

coefficient is: $\alpha = -\frac{1}{2d} \ln(\xi_{ref} \frac{A_d}{A_0})$ and A_d are the

ultrasonic amplitudes at $x=0$ and after having cover a distance $x=d$. ξ_{ref} Ratio of the acoustic impedances of the extreme mediums.

2. SLURRY CLAY PROPERTIES

Slurries of clay properties include mixture density (ρ_m), mass fraction (C_m) or volume fraction (C_v), mean size particle $\langle a \rangle$ and bulk density of the solid (ρ_c). The mass of the slurry (M_m) is: $M_m = V_f + M_c$, where V_f is the mass of fluid and M_c is the volume of the particulate of clay. The volume of slurry (V_m) is: $V_m = V_f + V_c$, where V_f is the volume of fluid and V_c is the volume of the particulate. The mass fraction of slurry is determined by the ratio

$$C_m = \frac{M_c}{(M_c + M_f)} = \frac{\rho_c (\rho_m - \rho_f)}{\rho_m (\rho_c - \rho_f)} \quad (1)$$

Where ρ_f is the fluid density. The corresponding volume fraction relationship is

$$C_v = \frac{C_m}{[C_m + S(1 - C_m)]} \quad (2)$$

Where S is the solids relative density (ρ_p/ρ_f) the volume fraction is proportional to the number of particles per unit volume (n). Suppose there are N particles in the total volume (V_T) and each particle occupies a given volume (V_p). The volume fraction is given by: $C_v = NV_p/V_T = nV_p$.

The average particle size of clay is 10,38 μm . The density of the slurry was obtained also by weighing a known volume of the sample. The density of the slurry was also obtained from the known weight percentage of the slurry, the density of clay (2600 kg/m^3), and the density of water. When the particulate is insoluble in the liquid, the relationship is given by using Eq. (3). For process control for a given type of particulate in the slurry, the instrument calibration provides the attenuation as a function of weight percentage. When the instrument operates in-line, the attenuation in turn is related to the weight percentage of solids in the slurry. An example of an in-line sensor for process control is shown in Figure 1.

$$\text{Wt}\% = \frac{\rho_w (\rho_s - \rho_w)}{\rho_s (\rho_c - \rho_w)} \times 100\% \quad (3)$$

Where ρ_w is the density of water, ρ_c is the density of the clay, and ρ_s is the density of the slurry.

3. DESCRIPTION OF THE EXPERIMENTAL DEVICE

The measuring apparatus is used in various manners often similar, made up of: a generator of pulse, an oscilloscope, a container and transducers (Figure 1), present the experimental device used in this work.

The experimental study is led in a parallelepipedic container (length=200mm, width=80mm, thickness=10mm). To obtain exploitable signals, it is necessary to be not obstructed by reflexions on the walls of the container. This last is equipped with system of precise positioning for the piezoelectric samples and sensors. This container is water filled whose density

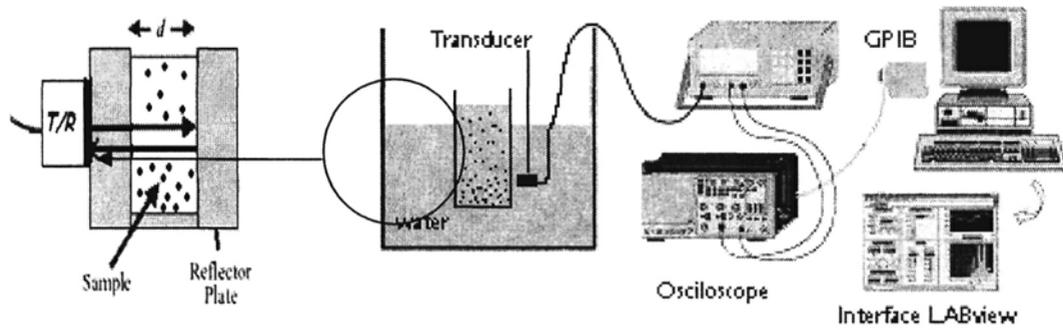


Fig. 1: Technique by reflexion

is $\rho_{eau} = 1000 \text{ kg/m}^3$ and in which the velocity of sound $c_{eau} = 1470 \text{ m/s}$, was measured at the ambient temperature of 298 K by [5, 6].

The experimental device consists of a generator of pulse SOFRANEL 5052pr which plays the part of receiving and transmitter which sends a very short pulse on the piezoelectric transducer Parametric type to broad band. The generated pulse is dispatched on the container of Plexiglas, containing water and various quantities of clays. The received signal is amplified and digitized by means of a numerical oscilloscope, and is sent towards a microcomputer, by an acquisition card GPIB (IEEE-488.2). The acquisition card is compatible with the graphic language LabVIEW; they are both built by National Instruments [7].

3.1 Real time Control of sedimentation clay particles under LabVIEW interface

We begin the control with the acquisition of the signals illustrated on the screen of the oscilloscope by an application under the graphic programming language LabVIEW. The developed interface makes it possible to specify the number of acquisitions wanted during the experiment and time separating two successive acquisitions. The LabVIEW program collects 50 signals each time and carries out an average to neutralize the interfering signals. The resulting signal represents for the user only one acquisition. An example of an experimental signal retrodiffused by the container which contains the sample is shown in Figure 2. This signal is composed of two echoes A_2 and A_4 shown successively from left to right on the figure. This signal is captured starting from the oscilloscope by the LabVIEW program

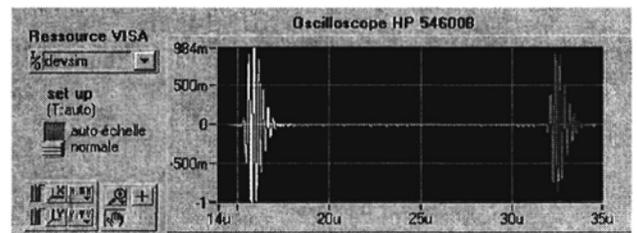


Fig. 2: Experimental signal retrodiffused by the container containing water and clay.

developed. We have noted that the water was characterized only by the echoes A_2 (reflexion on the interface between the second face of plate 1 and a mixture of (clay + water) locked up) and A_4 (reflexion with the interface between a mixture of (water + clay) and the first face of plate 2).

Then, the program separates the echoes by temporal windows by taking only the echo (A_2 and A_4) however the sites of echoes compared to time (X-coordinate) remain unchanged. The user has the possibility of choosing the first time manually the echo which will take a different color thereafter. The continuous spectra of phase of the echoes A_2 and A_4 are presented in Figure 3.

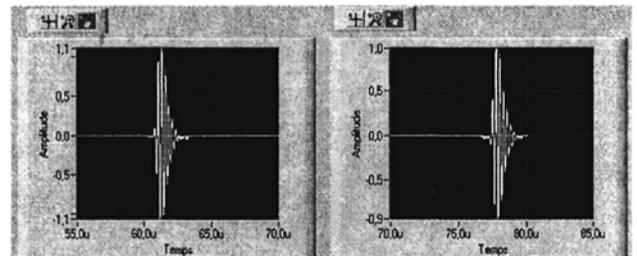


Fig. 3: Filtered signal of the A_2 echo and the A_4 echo.

The spectral amplitudes, of the echoes A_2 and A_4 , are determined by the application of the Fast Transform of Furrier to the signals of Figure 3. The spectra of amplitudes of the two echoes A_2 and A_4 are respectively presented by Figure 4.

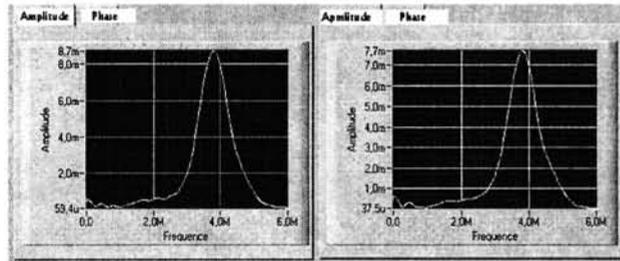


Fig. 4: Spectral amplitude of the A_2 echo and spectral Amplitude of the A_4 echo

3.2 Speed of sound in water clay mixture

The speed of sound of products is given by the following expression [5,8]

$$V_{product}(v) = \frac{\omega 2L}{\phi_{product / pg} - \phi_{pg / product}} \quad (4)$$

L is the thickness of the container, $\omega = 2\pi f$, $\phi_{pg / product}$ is the phase of the echo A_2 et $\phi_{product / pg}$ is the phase of the echo A_4 . The Transform of Fourier of the echo A_2 isolated by the program, gives the real part $R_2(\omega)$ and imaginary part $I_2(\omega)$, making possibility to calculate the phase according to the traditional relation:

$$\phi_{pg / product} = \arctg\left(\frac{I_2(\omega)}{R_2(\omega)}\right) \quad (5)$$

Whereas the Transform of Fourier of the A_4 echo insolated by the program, which, was developed in our laboratory, gives the real part $R_4(\omega)$ and imaginary part $I_4(\omega)$, making possibility to calculate the phase $\phi_{product / pg}$:

$$\phi_{product / pg} = \arctg\left(\frac{I_4(\omega)}{R_4(\omega)}\right) \quad (6)$$

In the calculation of the two expressions (5) and (6), we obtain two spectra whose phase varies between $-\pi/2$ and $\pi/2$. We conceived a LabVIEW interface with an algorithm allowing to unroll the phases in continuous form. Using the algorithm of the phases, and the developed LabVIEW interface, we have determined the speed of sound of product locked up according to the frequency.

3.3 Attenuation of sound in water clay mixture

The attenuation of sound in water clay mixture is given by the following equation: [6,9,10].

$$\alpha(v) = -\frac{1}{2L} \ln\left(\frac{A_4}{A_2} \xi_{ref}\right) \quad (7)$$

With $Z = \rho_{pg} \cdot C_{pg}$, $\xi_{ref} = \frac{(Z_{pg} + Z_{eau+arg})^2}{4Z_{pg} Z_{eau+arg}}$, and $Z_{eau+arg} = \rho_{eau+arg} \cdot C_{eau+arg}$, Z_{pg} is the acoustic impedance in the Plexiglas, $Z_{eau+arg}$ is the acoustic impedance in water clay mixture.

4. RESULTS AND DISCUSSION

The generated pulse is dispatched towards the container containing water clay mixture with various concentrations, after a sufficient mixture, which ensures the average distribution of the particles by volume element, the suspension is leaved sediment.

The curves obtained in Figure 5 shows that the water containing clay with concentrations (5g/L-10g/L, 20g/L, 30g/L and 40g/L) are different, and start to show stages two minutes after the beginning of the experiment. The curves obtained in Figure 6 shows that the concentrations (5g/L-10g/L, and 15g/L of clay) are almost the same ones. On the other hand, at high concentrations of clay, there is a fall of attenuation at 11 minutes after the beginning of experiment, due to the effect of fast decantation.

Data, shown in Table 1, were obtained for the attenuation of ultrasound through clay slurries, having a weight percentage of 1%, 2%, 10%, and 20%.

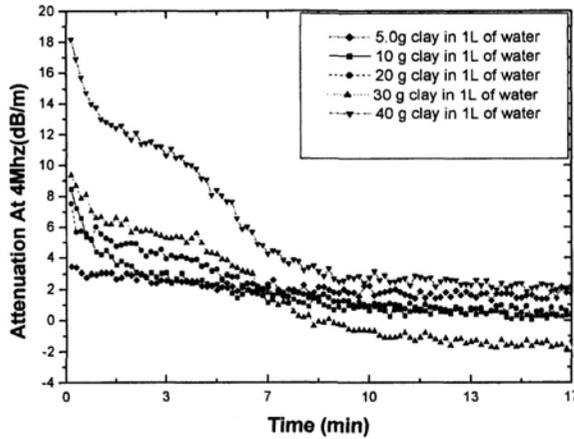


Fig. 5: Attenuation curves in water containing various concentrations of (Transducer used 4 MHz)

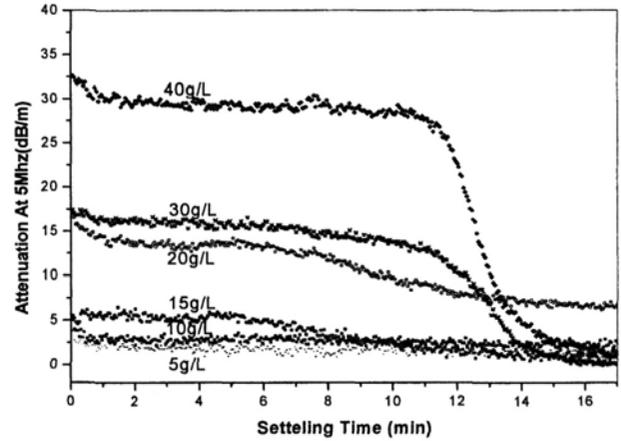


Fig. 6: Attenuation curves in water containing various concentrations of clay (Transducer used 5 MHz)

Table 1
Data obtained for clay slurries

Slurry	Velocity (m/s)	Independent density Measurement (Kg/m ³)	Sensor density (Kg/m ³)	Density obtained from Weight% (Kg/m ³)	Z _{liq} (Kg/m ² s)
Water	1489	998	993	-----	1486022
1% clay	1482	1003	1016	1004	1486446
2% clay	1487	1009	1032	1010	1500383
10% clay	1484	1061	1035	1064	1574524
20% clay	1489	1087	1087	1098	1618543

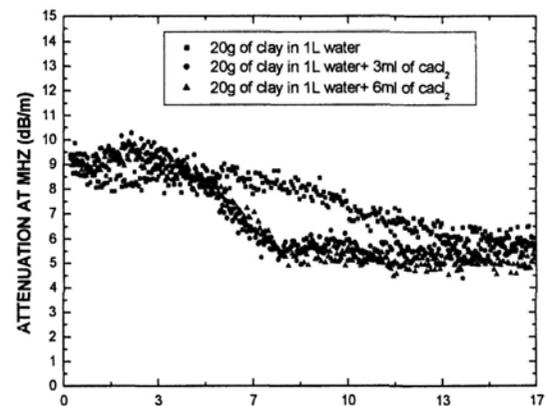
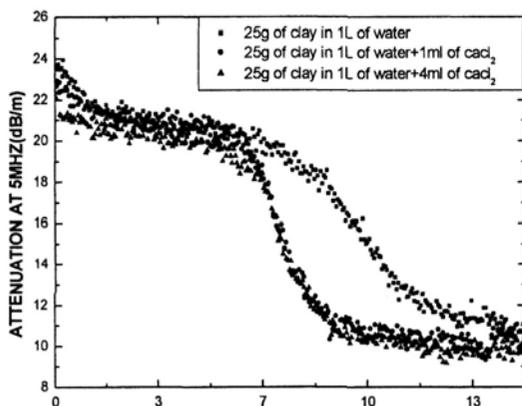


Fig. 7: Curves of sound attenuation in water and clay mixture with different concentration of the coagulant CaCl₂

The addition of the chemical component CaCl_2 on the two clay solutions, which correspond to the concentrations 20g/L and 25g/L causes a remarkable variation in attenuation after 5 minutes at the beginning of the experiment, which shows that the sedimentation rate increased after the clay particles reacted with the CaCl_2 component, as shown in Figure 7.

5. THEORY

The goal with this section is to derive an expression for the attenuation of sound that passes through a suspension of small solid particles of clay and water. For a sound wave that travels in pure water we can consider the water to be lossless. If, however, an object such as a small particle obstructs the wave, the wave will lose energy, and the most dominant loss process is scattering. In a suspension with many particles the scattering will repeat itself many times. When the incoming plane wave interacts with a particle, the sound is scattered in all directions. Assume that all sound energy scattered away from the receiving transducer is lost.

Then, if all particles are of equal size and they all have the same mechanical properties the energy loss, $d\xi$, caused by N particles can be written as

$$d\xi = -N \xi_{réfléchi} \tag{8}$$

Where $d\xi$ is the back scattered energy from each particle and N is the number of particles in the path of the wave. In order to proceed we must determine the number of particles in the path of the wave, and the scattered energy from each particle. The mass fraction, C_m , of particles in the suspension is

$$C_m = \frac{N m_{particle}}{m_{liquid} + N m_{particle}} \approx \frac{N m_{particle}}{m_{liquid}} \tag{9}$$

Where $m_{particle}$ is the mass of one particle and m_{liquid} is the mass of the liquid phase of the suspension is mass of the liquid phase of the suspension. For a control volume with geometry as shown in Figure 8.

Having width $w=80\text{mm}$, height $h=200\text{mm}$, and length $dx=10\text{mm}$ the mass of fluid inside the control volume is given by

$$m_{liquid} = \rho_{liquid} V_{liquid} = \rho_{liquid} \times (w \times h \times dx) \tag{10}$$

and the mass of a particle is

$$m_{particle} = \rho_p \frac{4}{3} \pi \langle a \rangle^3 \tag{11}$$

Where ρ_{liquid} and $\rho_{particles}$ is the densities of the liquid and the particles, respectively, and $\langle a \rangle$ is the mean particle radius. From this we can estimate the number of particles as

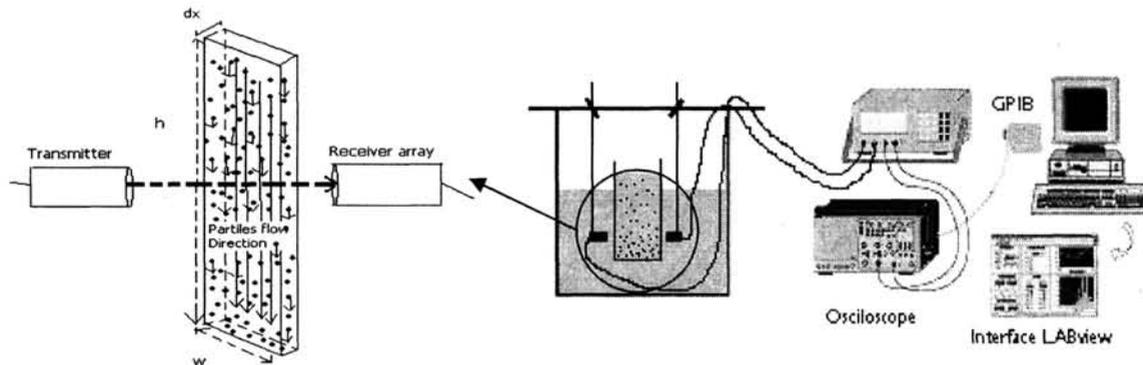


Fig. 8: Transmitter and receiver array used in the measurements, with notations used in the derivation of the attenuation coefficient. (Technique by transmission).

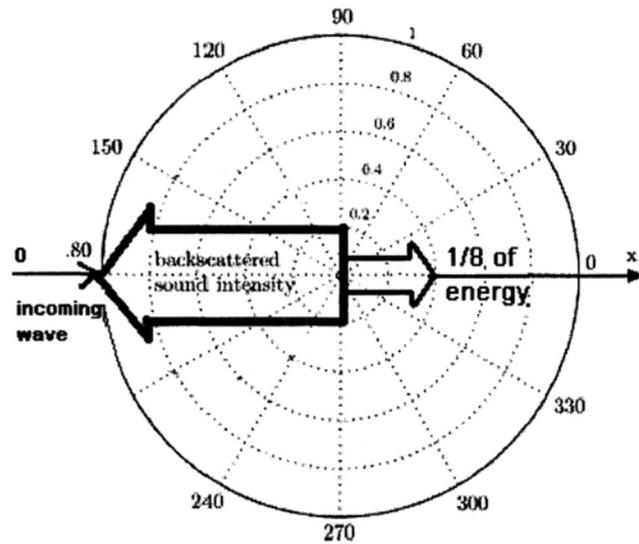


Fig. 9: Scattering from a rigid particle when a plane wave is incident from the left in the picture ($k_a=0,3$)/12,13/

$$N \approx \frac{m_{eau}}{m_{particle}} = C_m \frac{3}{4} \frac{\rho_{eau} w h}{\rho_{particle} \pi \langle a \rangle^3} \quad (12)$$

Now, when we have an estimation of the number of particles inside the control volume, we need to determine how each of these clay particles scatters the incoming sound wave. It should be noted, however, that if particles conglomerate, this will change the number of particles. It can be shown /11/ that when a plane wave of intensity I encounters an incompressible rigid particles, the scattered intensity I_s is given by

$$\frac{I_s}{I} = \frac{16\pi^4 f^4}{9 C^4 r^2} \langle a \rangle^6 (1 - 3 \cos \theta)^2 \quad (13)$$

Where F is the frequency, c is the speed of sound in the surrounding medium, r and h are the radius and angle coordinates of a polar coordinate system, respectively.

This expression is valid for wavelengths such that $k_a \ll 1$, where a is particle radius and k is the wave number. Figure 9 shows the pattern of the scattered intensity. Even though Eq. (13) is described by the polar coordinates r and h , it may be valid in three dimensions, because of symmetry around the x -axis. As expected and as Figure 9 shows, most of the energy is backscattered, i.e. scattered in the opposite direction of

the incoming sound wave. In fact only 1/8 of the energy is scattered forward (to the right in Figure 9. The same ratio is true as the long wavelength approximation holds ($k_a \ll 1$). The total backscattered power is found by integration of Eq. (13) over a half sphere with origin at the same position as the particle in question that is:

$$\xi_{\text{rétrodiffusé}} = \int_{\frac{\pi}{2}}^{\pi} I_s(r, \theta) 2\pi r^2 \sin \theta d\theta \quad (14)$$

Where the frequency has been replaced by wavelength $\lambda = c/f$. For the control volume in Figure 8, the intensity of the incoming wave is related to the energy by Eq. (14), Where ξ is the incoming plane wave, at position x , and $D=16\text{mm}$ is the diameter of the transmitter transducer.

$$I = \frac{\xi}{\pi \left(\frac{D}{2}\right)^2} = \frac{4 \xi}{\pi D^2} \quad (15)$$

The backscattered energy now becomes

$$\xi_{\text{rétrodiffusé}} = \frac{16\pi^4 f^4}{9 C^4} \langle a \rangle^6 I \int_{\frac{\pi}{2}}^{\pi} (1 - 3 \cos \theta)^2 2\pi \sin \theta d\theta \quad (16)$$

$$\xi_{retrodiffusé} = \frac{114}{9} \frac{\pi^5 \langle a \rangle^6}{\lambda^4} I = \frac{152}{3} \frac{\pi^4 \langle a \rangle^6}{\lambda^4 D^2} \xi \quad (17)$$

The use of Eq. (17) together with Eq. (12) leads to an ordinal differential equation for the energy loss of the sound wave.

$$\frac{d\xi_{ref}}{\xi} = -\frac{152}{3} \frac{\pi^4 \langle a \rangle^6}{\lambda^4 D^2} \times C_m \frac{3}{4} \frac{\rho_{eau} w h}{\rho_{particle} \pi \langle a \rangle^3} \quad (18)$$

$$\frac{d\xi_{ref}}{\xi} = -A \frac{\langle a \rangle^3}{\lambda^4} \frac{\rho_{eau}}{\rho_{particle}} C_m dx \quad (19)$$

Where the constant $A = 38\pi^3 \frac{wh}{D^2}$ Integration of both sides gives

$$\int_{\xi_0}^{\xi} \frac{d \bar{\xi}_{ref}}{\bar{\xi}} = - \int_0^x A \frac{\langle a \rangle^3}{\lambda^4} \frac{\rho_{eau}}{\rho_{particle}} C_m \bar{dx} \quad (20)$$

Where $\bar{\xi}$ and \bar{x} are integration variables, and ξ_0 is transmitted energy at position $x=0$. After integration we obtain;

$$\frac{\xi}{\xi_0} = e^{-C_m A \frac{\langle a \rangle^3}{\lambda^4} \frac{\rho_{eau}}{\rho_{particle}} x} = e^{-\alpha x} \quad (21)$$

Where the attenuation of sound in the suspension is

$$\alpha = C_m A \frac{\langle a \rangle^3}{\lambda^4} \frac{\rho_{eau}}{\rho_{particle}} \quad (22)$$

It is the attenuation coefficient due to backscattering. As expected, the attenuation in Eq. (21) depends on the mass fraction of particles and the propagation distance of the wave. For a measurement system where the distance between the transducers is constant (see Figure 8) the measured attenuation coefficient, α , will be proportional to the mass fraction, C_m , alone. The expression for α in Eq. (22) is valid as long as the mass fraction of particles is low so that we can assume

that there is no multiple scattering. Furthermore, in the model we assume a constant mean particle radius, $\langle a \rangle$, and constant wavelength of the sound.

6. CONCLUSION

At high concentrations of clay, there is a fall of attenuation at certain moment of the decantation, due to the effect of fast decantation. And there are appearances of plates at the beginning of experiment. It shows that the behavior is not the same for the concentrated slurries and no concentrated in clay. Each concentration is characterized by a clean acoustic signature. The experimental results show that the attenuation of sound due to the particles of clay varies almost linearly with mass fraction of clay, (see Figure 10).

The addition of 0.9% of $CaCl_2$ each mixture of water and clay makes it possible to accelerate the sedimentation test. We have presented a simple

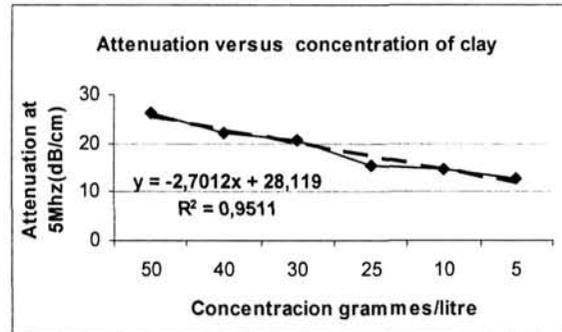


Fig. 10: The attenuation coefficient varies linearly with particle mass fraction. Dotted: experiment; line: model

theoretical model for the coefficient of excess attenuation, α , of pulsed ultrasound due to the presence of particles.

The model predicts that the attenuation coefficient varies linearly with particle mass fraction. The ultrasounds waves are sensitive to the small quantities of clays. We can also control the addition of the chemicals components used to recover clays in suspension in the basins of stopping water.

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