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# Effective Medium Model for Ultrasonic Attenuation Due to the Thermo-Elastic Effect in Concentrated Emulsions

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**Abstract.** — In this article, we deal with the propagation of ultrasonic waves in monodisperse concentrated oil-in-water emulsions. Using the approximation of Isakovich, we propose two different models, a mirror model and a core-shell model, aiming to describe the temperature field in the dense medium and to supply a correct expression of the ultrasonic wave vector. The comparison between experimental data and theoretical models shows that the core-shell model leads to a very accurate description of the ultrasonic attenuation, in a wide range of frequencies and concentrations, in the case where the thermo-elastic effect, due to the scattering of thermal waves by the particles, is the dominant loss mechanism.

## 1. Introduction

Application of ultrasonic absorption measurements to the determination of the size of particles suspended in a fluid is being given more and more attention [1]. The major advantage of this technique resides in the fact that it is non destructive and can be employed with concentrated systems. The theory of the propagation of ultrasonic waves in dilute suspensions is now well-established [2–6], but recent studies [7,8] have shown that, at concentrations higher than about 10%, classical models fail to explain quantitatively the ultrasonic attenuation, particularly for small droplets at low frequencies for which attenuations much smaller than the predicted values are observed. This discrepancy has been attributed [7,8] to the interactions between particles which set up in these circumstances.

The propagation of ultrasonic waves is controlled, in the case of emulsions at moderate frequencies, by essentially one mechanism named the thermo-elastic effect, related to the irreversibility of heat exchanges between the two phases. The theory of this effect has been established in 1948 by Isakovich [9] in the case of dilute suspensions. An attempt for modeling concentrated suspensions has been proposed by Fukumoto *et al.* [10] who have developed a model in which the particles occupy the nodes of a periodic network, but the complexity of the calculations involved seems to limit the practical interest of their approach.

In this paper we propose a modelization of the propagation of ultrasonic waves in concentrated emulsions by using approximations which lead to manageable formulations. The first

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one consists in approximating the zero heat flux surface around each droplet by a spherical mirror. The second is a core-shell approximation, where the core is the oil droplet, the shell is the layer of water in contact with the droplet and outside the shell lies an effective medium. Both these models use no adjustable parameter, it will be seen that the core-shell model offers very accurate results. We will first recall the method of Isakovitch for calculating the ultrasonic propagation in an inhomogeneous medium with heat transfer. We will then describe the mirror and the core-shell models by using a generalization of the method of Isakovitch. Finally we will present a comparison between our predictions and experimental results.

## 2. Theory

2.1. ISAKOVICH APPROACH OF THERMAL SCATTERING. — In order to modelize the part of the sound absorption due to thermal dissipation in an heterogeneous medium Isakovitch [9] has proposed a general method which can be applied to any medium as soon as the pressure wavelength is large compared to the typical heterogeneity lengthscale in the sample. In fact this approximation facilitates the computation of the spatial dependence of the heat fluxes induced in the medium by the modulation of the pressure.

The approach of Isakovitch has been fully described by Fukumoto *et al.* [11]. We recall here the essential points. By writing the conservation of momentum, mass, and energy in the medium, two linearized expressions are obtained [11]:

$$\frac{\partial \delta T}{\partial t} - \frac{1}{\rho C_p} \nabla \cdot (\kappa \nabla \delta T) - \frac{T\beta}{\rho C_p} \frac{\partial \delta P}{\partial t} = 0 \quad (1)$$

$$\chi_T \frac{\partial^2 \delta P}{\partial t^2} - \nabla \cdot \left( \frac{1}{\rho} \nabla \delta P \right) - \beta \frac{\partial^2 \delta T}{\partial t^2} = 0 \quad (2)$$

where  $\delta P$  is the pressure fluctuation,  $T$  and  $\delta T$  the temperature and its fluctuation.  $\rho$  is the density,  $\beta$  is the thermal expansion coefficient,  $\chi_T$  the isothermal compressibility,  $\kappa$  the thermal conductivity and  $C_p$  the specific heat at constant pressure. The first equation is just the equation of heat propagation, but where the last term expresses the fact that the pressure variations act as heat sources. The second equation describes the propagation of the pressure wave in a medium whose compressibility is affected by the modulation of the temperature. The last terms of these two equations are conjugate and describe the coupling between the temperature field and the pressure wave. At first view, solving this system is difficult as each thermodynamical variable depends on the position; the approximation of Isakovitch is helpful because the local variations of the pressure are going to be neglected.

Consider now a periodic pressure wave and a periodic temperature field of frequency  $\omega$ , the pressure field is assumed to be homogeneous and such that it can be approximated by:

$$\delta P = \delta P_0 e^{-i(\omega t - \mathbf{k}\mathbf{r})} \quad (3)$$

where  $\mathbf{k}$  is the wave vector. The temperature field, for the moment, is simply assumed to be periodic:

$$\delta T = \delta T_0(\mathbf{r}) e^{i\omega t} \quad (4)$$

Equation (2) is averaged in order to get the propagation equation for the pressure wave:

$$k^2 = \omega^2 \langle \rho \rangle \left\langle \chi_T - \beta \frac{\delta T_0}{\delta P_0} \right\rangle \quad (5)$$

where  $\langle \dots \rangle$  stands for spatial average. The main assumption is that the pressure field is not affected at small scale by the thermal field. This is true as long as the sound wavelength is

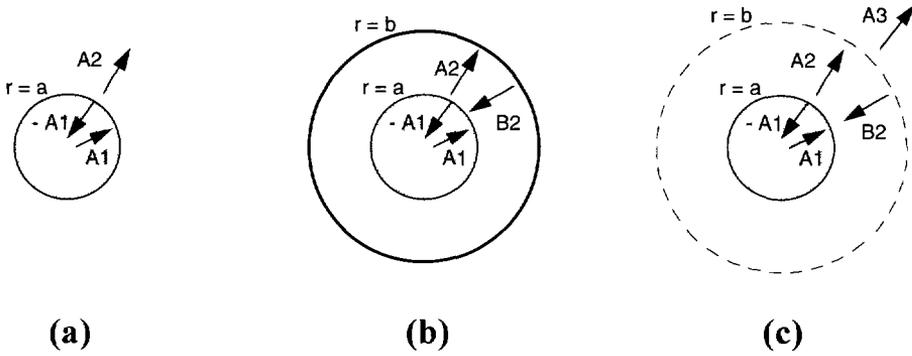


Fig. 1. — (a) Model of Isakovich: an isolated oil droplet exchanges heat with the water outside. (b) The mirror approximation: the droplet is placed at the center of a spherical mirror simulating the heat radiated by neighbor droplets. (c) The core-shell model: the oil core is at the center of a water shell surrounded by an effective medium.

large compared to the thermal wavelength (in liquids this condition is verified up to the GHz range).

Using equation (1), the following relation for the acoustic wave vector is then obtained:

$$k^2 = \omega^2 \rho_0 \frac{1}{V} \int_V \left[ \left( \chi_T - \frac{T\beta^2}{\rho C_p} \right) - \frac{i\beta}{\omega \rho C_p} \nabla \cdot \left[ \frac{\kappa \nabla \delta T_0(\mathbf{r})}{\delta P_0} \right] \right] dV \quad (6)$$

where  $V$  is the volume of the system,  $T$  the temperature at rest and  $\rho_0$  the average density.

This expression allows to calculate the propagation of the ultrasonic wave in a dilute suspension of particles, in the case where only thermal waves are scattered.

2.2. SOUND PROPAGATION IN DILUTE SYSTEMS SPHERICAL PARTICLES. — Consider a spherical particle of type 1 and of radius  $a$ , embedded in a matrix of type 2 (Fig. 1a). The subscripts 1 and 2 will stand for the properties of medium 1 and 2 respectively.

In equation (6) the first term in the integral is the adiabatic compressibility of each medium, so that the integration of this quantity over the whole volume is simply the average adiabatic compressibility  $\chi_0$ . The average density and compressibility are the volume averages of the properties of the components, given by:

$$\rho_0 = \phi \rho_1 + (1 - \phi) \rho_2, \quad \chi_0 = \phi \chi_1 + (1 - \phi) \chi_2. \quad (7)$$

These two quantities lead to the limiting average velocity:

$$c_0 = \frac{1}{(\rho_0 \chi_0)^{1/2}} \quad (8)$$

which will be shown to be the limit of the velocity at very high frequencies. Equation (6) can now be rewritten as:

$$k^2 = \frac{\omega^2}{c_0^2} \left[ 1 - \frac{i}{\omega \chi_0} \frac{1}{V} \int_V \frac{\beta}{\rho C_p} \nabla \cdot \left[ \frac{\kappa \nabla \delta T_0(\mathbf{r})}{\delta P_0} \right] \right] dV. \quad (9)$$

Assuming that the wavelength of the compression wave is large compared to the size of the particle, so that the pressure can be considered as uniform in an around the particle,

the thermal field in each medium is then obtained from equations (1), (3) and (4). In spherical coordinates, with the center of the sphere at the origin, the thermal field in each medium will be given by the following expressions:

$$\delta T_1 = \left[ \frac{T\beta_1}{\rho_1 C_{p1}} + \frac{A_1}{r} \sinh(n_1 r) \right] \delta P_0 e^{2\omega t} \quad \text{for } r \leq a \quad (10)$$

$$\delta T_2 = \left[ \frac{T\beta_2}{\rho_2 C_{p2}} + \frac{A_2}{r} \exp(-n_2 r) \right] \delta P_0 e^{2\omega t} \quad \text{for } r \geq a \quad (11)$$

where  $n_j = (1-i)(\omega\rho_j C_{pj}/2\kappa_j)^{1/2}$  is the wave number of the diffusive thermal wave travelling in medium  $j$ , solution of equation (1) for a periodic excitation. The form of these solutions results from the fact that the temperature fluctuation must be finite at the center of the particle, and must vanish at large distances.

The amplitudes  $A_1$  and  $A_2$  are obtained by applying the boundary conditions which express the continuity, at the surface of the particle, of both the temperature and the thermal flux; these conditions read:

$$\delta T_1|_{r=a} = \delta T_2|_{r=a} \quad \text{and} \quad \kappa_1 \frac{\partial \delta T_1}{\partial r} \Big|_{r=a} = \kappa_2 \frac{\partial \delta T_2}{\partial r} \Big|_{r=a} \quad (12)$$

The volume integral in equation (9) is evaluated at first for a single particle, by transforming the volume integral of the divergence into a surface integral by use of Gauss's theorem, taking into account equation (12) and noting that the heat flux vanishes at infinity:

$$\int_0^a (1) + \int_a^\infty (2) = \int_0^a (1) - (2) + \int_0^\infty (2) = 4\pi a^2 \left( \frac{\beta_1}{\rho_1 C_{p1}} - \frac{\beta_2}{\rho_2 C_{p2}} \right) \kappa_1 \frac{\partial \delta T_1}{\partial r} \Big|_{r=a} \quad (13)$$

where  $(j)$  stands for the integrand in medium  $j$ .

An approximation has then to be made for summing over the  $N$  particles which are found in the volume  $V$ : it is supposed that, at a distance comparable to the separation between particles, the heat flux has sufficiently decreased so that it may be neglected. This approximation is precisely the reason why the model fails to describe the ultrasonic behavior of concentrated suspensions.

Noting that the volume fraction of the droplets is given by:

$$\phi = \frac{N}{V} \frac{4}{3} \pi a^3 \quad (14)$$

the result of Isakovich is finally obtained:

$$k(\omega) = \frac{\omega}{c_0} \left[ 1 + i \frac{3\phi}{\omega a^2} \frac{T}{\chi_0} \left( \frac{\beta_1}{\rho_1 C_{p1}} - \frac{\beta_2}{\rho_2 C_{p2}} \right)^2 A \right]^{1/2} \quad (15)$$

where

$$A = \left[ \frac{1}{\kappa_1 [n_1 a / \tanh(n_1 a) - 1]} + \frac{1}{\kappa_2 (1 + n_2 a)} \right]^{-1}$$

The real part  $\text{Re}(k)$  and the imaginary part  $\text{Im}(k)$  of the wave vector lead to the celerity of the wave  $c(\omega)$ , to the absorption coefficient  $\alpha(\omega)$ , and to the loss per cycle  $\alpha \cdot \lambda(\omega)$ :

$$\begin{aligned} c(\omega) &= \omega / \text{Re}(k) \\ \alpha(\omega) &= \text{Im}(k) \\ \alpha \cdot \lambda(\omega) &= 2\pi \text{Im}(k) / \text{Re}(k). \end{aligned} \quad (16)$$

At low frequencies, the range of the thermal wave is large compared to the size of the particles, and the loss per cycle scales in  $\omega$ , as in any other relaxation process. At high frequencies, where the range of the thermal wave is small compared to the size of the particles, the dissipation occurs within thin layers surrounding the interface. The loss per cycle scales then in  $\omega^{-1/2}$  as the range of the thermal wave. Between these two regimes the loss per cycle goes through a maximum at a frequency for which the product  $n_1 \cdot a$  is close to unity, that is where the range of the thermal wave is comparable to the size of the droplet. It is interesting to remark that in the above propagation equation the loss per cycle appears as a function of a reduced variable equal to the product of the frequency by the square of radius of the particle [7, 8].

This analysis offers excellent results as far as the distance between particles is large enough, so that the thermal field (Eqs. (10, 11)) is not deeply modified by the presence of the surrounding particles. In order to describe the temperature field in a concentrated solution, we have tried, in a first time, a "mirror" approximation.

**2.3. THE MIRROR APPROXIMATION.** — Let us consider a crystalline array of spherical particles, as in [10]. Each plane equidistant between two neighboring particles is a symmetry plane such that there are as many heat sources on one side of this plane as on the other. Thus, if we assume that the pressure wavelength is large compared to the distance between particles, all these heat sources will be nearly in phase, and the heat flux will vanish on each of the symmetry planes. The Wigner-Seitz cell that these planes build around each particle is a polyhedron where the heat flux is equal to zero, so that this polyhedron may be considered as a mirror which reflects the heat flux. As it is quite intricate to compute directly the heat flux in a polyhedron, we will approximate the polyhedron by a sphere (Fig. 1b) whose radius  $b$  will be equal to half the mean distance between particles.

We have now to consider one more wave: the thermal wave reflected by the mirror. For computing the amplitude  $B_2$  of this wave we take advantage of one more condition: the thermal flux must vanish on the mirror. The wave number of the acoustic wave is thus calculated by taking into account the perturbation of the temperature field around the particle due to the reflected thermal wave. An expression similar to equation (15) is obtained, but in which the quantity  $A$  is somewhat more complicated:

$$A = \left[ \frac{1}{\kappa_1 [n_1 a / \tanh(n_1 a) - 1]} + \frac{1}{\kappa_2 [1 + n_2 a (1 + B) / (1 - B)]} \right]^{-1} \quad (17)$$

where

$$B = \frac{1 + n_2 b}{1 - n_2 b} e^{-2n_2(b-a)}$$

This model does predict a decrease of the attenuation at low frequency, where the range of the thermal wave is large compared to the radius of the mirror so that the thermal gradient around the droplet is lowered. At high frequency, as the thermal wavelength goes to zero, the classical result of Isakovich is found again.

But for obvious geometrical reasons, this model is limited to concentrations below the close-packing, for which the thermal exchanges would be blocked by the mirror! In fact it appears that the model underestimates the attenuation in the whole concentration domain. This underestimation is probably related to the replacement of a polyhedron by a sphere. In a sphere, there is only one typical length for a field with a radial symmetry: namely the radius. But in a polyhedron, as the wave is reflected by plane surfaces linked by edges, smaller typical lengths have to be taken into account which result in larger temperature gradients. Equivalently it could be said that the mirror reflects the thermal wave too strongly, and that a "porous" sphere could improve the result. We will now present such an approach.

2.4. THE CORE-SHELL MODEL. — We shall now assume that the space next to the droplet is occupied by pure water, while an effective medium occupies the rest of the space, as sketched Figure 1c. The boundary between the water and the effective medium is assumed to be a sphere whose radius  $b$  is such that the oil volume fraction is the same inside the sphere and in the effective medium. The value of  $b$  is then given by:

$$b = \frac{a}{\phi^{1/3}}. \quad (18)$$

It is to be emphasized that this model does not require any other supposition or adjustable parameter. A similar approximation has been used by several authors [12–14] in other mechanical situations. This model should describe correctly what happens at low frequency, where the effective medium approximation has been shown to be adequate [8], as well as at high frequency, where the range of the thermal wave is smaller than the distance between droplets. The effective medium, denoted by the subscript 3, is described by the following thermodynamical constants [8]:

$$\begin{aligned} \rho_3 &= \phi\rho_1 + (1 - \phi)\rho_2 \\ \rho_3 C_{p3} &= \phi\rho_1 C_{p1} + (1 - \phi)\rho_2 C_{p2} \\ \beta_3 &= \phi\beta_1 + (1 - \phi)\beta_2 \end{aligned} \quad (19)$$

which are the volume averages of the properties of the components, obtained as in equation (7) ( $\rho_3$  is the same quantity as  $\rho_0$ ). As regards the thermal conductivity, which cannot simply be obtained as a volume average, use has been made of the expression given by Torquato [15] for a random dispersion of hard spheres:

$$\kappa_3 = \kappa_2 \frac{1 + 2\phi\gamma - 2(1 - \phi)\zeta\gamma^2}{1 - \phi\gamma - 2(1 - \phi)\zeta\gamma^2} \quad (20)$$

$$\text{where } \gamma = \frac{\kappa_1 - \kappa_2}{\kappa_1 + 2\kappa_2} \quad \text{and} \quad \zeta = 0.21068\phi - 0.04693\phi^2.$$

In the three successive regions, the temperature field is then given by the following expressions:

- for  $r < a$ , inside the droplet itself:

$$\delta T_1 = \left[ \frac{T\beta_1}{\rho_1 C_{p1}} + \frac{A_1}{r} \sinh(n_1 r) \right] \delta P \quad (21)$$

- for  $a < r < b$ , in the water shell:

$$\delta T_2 = \left[ \frac{T\beta_2}{\rho_2 C_{p2}} + \frac{A_2}{r} \exp(-n_2 r) + \frac{B_2}{r} \exp(n_2 r) \right] \delta P \quad (22)$$

- for  $r > b$ , in the effective medium outside the shell:

$$\delta T_3 = \left[ \frac{T\beta_3}{\rho_3 C_{p3}} + \frac{A_3}{r} \exp(-n_3 r) \right] \delta P. \quad (23)$$

The values of the amplitudes  $A_i$  and  $B_i$  are worked out by using the boundary conditions on the temperatures and the heat fluxes at both interfaces, for  $r = a$  and for  $r = b$ . By use of

relation (9), we finally get a dispersion relation for the ultrasonic wave similar to equation (15), but in which the quantity  $A$  is now given by:

$$A = \kappa_1 \kappa_2 \frac{n_1 a - \tanh(n_1 a)}{E \cdot C + F \cdot D} \left[ 2\kappa_3 \frac{g_2 - g_3}{g_1 - g_2} n_2 b (n_3 b + 1) + C(1 + n_2 a) + D(1 - n_2 a) \right]$$

where

$$g_j = \beta_j / (\rho_j C_{p,j}) \quad (24)$$

$$\begin{aligned} C &= e^{n_2(b-a)} [\kappa_2(n_2 b - 1) + \kappa_3(n_3 b + 1)] \\ D &= e^{-n_2(b-a)} [\kappa_2(n_2 b + 1) - \kappa_3(n_3 b + 1)] \\ E &= \kappa_1 n_1 a + [\kappa_2(n_2 a + 1) - \kappa_1] \tanh(n_1 a) \\ F &= \kappa_1 n_1 a - [\kappa_2(n_2 a - 1) + \kappa_1] \tanh(n_1 a). \end{aligned}$$

It is easily verified that the above expression reduces to equation (15) both for high frequencies and for low concentrations (in these two cases the term  $C$  diverges because  $n_2 \cdot b$  goes to infinity). In fact in these conditions the reflected thermal wave is entirely attenuated before it can reach the droplet so that no perturbation of the temperature field occurs and the formulation of Isakovitch is perfectly valid.

It is interesting to notice that, here also, the losses appear as a function of the reduced variable  $f \cdot a^2$ , which offers a convenient way of presenting the results pertaining to different classes of size on a single plot.

Concerning the above expression, an important point has to be made clear: we have deliberately omitted one term in the derivation of equation (24). We have to integrate the divergence of the heat flux over the whole volume, this integral can be symbolically decomposed, as in relation (13):

$$\int_0^a (1) + \int_0^b (2) + \int_0^\infty (3) \quad (25)$$

where (1), (2) and (3) stand for the integrand in each successive medium:

- the first term is exactly what we need, namely the losses due to the oil core in a thermal field which is perturbed by the thermal wave reflected from the outside;
- the last term is equal to zero because the heat flux vanishes at large distances;
- the second term must be discarded as it represents the virtual contribution of a suspension of water droplets in the effective medium, which would interfere with the first term and result in an overestimation of the losses if it was taken into account. In other words this contribution is already taken into account when the contribution of the first term is summed over all the particles.

Figure 2 presents a comparison of the results of the core-shell model with the model of Isakovitch for a typical emulsion. The variations of the loss per cycle  $\alpha \cdot \lambda$  are plotted as a function of the volume fraction  $\phi$  for different frequencies. In order to clarify the presentation the vertical scale has been adapted for each frequency in such a way that all curves present identical slopes at the origin. The frequencies indicated are relative to the frequency for which the range of the thermal wave is comparable to the size of the droplet.

As expected, the fundamental result is that the losses start to lessen, compared to the predictions of Isakovitch, above a threshold concentration which increases with the frequency.

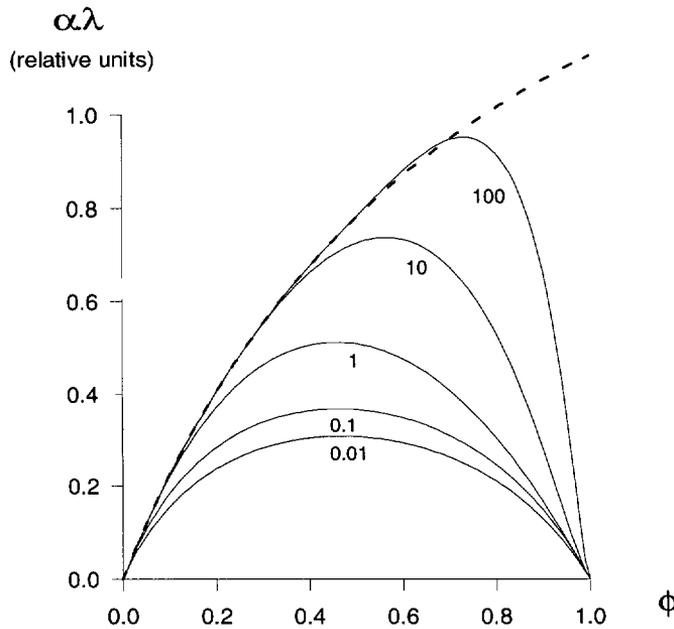


Fig. 2. — The predictions of the core-shell model (solid lines) are compared to the model of Isakovich (dashed line). The loss per cycle is plotted as a function of the oil volume fraction for a number of frequencies (for an easier comparison the vertical scale is adapted in such a way that all curves present the same slope at the origin, in this representation the curves predicted by the model of Isakovich have essentially the same shape for all frequencies). It can be seen that the departure between the two models occurs for a volume fraction which is lower at low frequencies, where thermal waves can propagate at large distances, than for high frequencies, where thermal interactions vanish.

### 3. Experimental Verification

In order to confirm the validity of our model two series of ultrasonic experiments have been performed. We have studied two kinds of emulsions, made of a polysiloxane oil and of n-hexadecane respectively. These oil/water emulsions are prepared by using a magnetostrictive homogenizer, they are stabilized with sodium dodecyl-sulfate as surfactant, and finally they are fractionated by using the selective creaming method proposed by Bibette [16]. The samples present a very narrow size distribution, so that the results can be adequately compared to the theory. The ultrasonic attenuation has been measured using a modified version of the acoustic resonator described by Eggers [17], between 500 kHz and 10 Mhz at room temperature. All the experimental details have been reported in a previous publication [8]. The thermo-physical constants of the components needed for the modeling are listed in Table I.

The ultrasonic absorption spectra obtained for both type of emulsions are presented in Figures 3 and 4, along with the predictions of the model of Isakovich and of our core-shell model. For each concentration the data pertaining to the different classes of size are presented as a single spectrum by use of the  $f \cdot a^2$  representation. The losses due to the intrinsic absorption of the constituents have been subtracted from the measurements, so that the quantity which is shown is the excess absorption due to the thermo-elastic losses. It can be seen that the core-shell model is successful in the whole size and frequency range, up to concentrations of 50%, for which the model of Isakovich is seriously mistaken. In fact an estimation of the particle

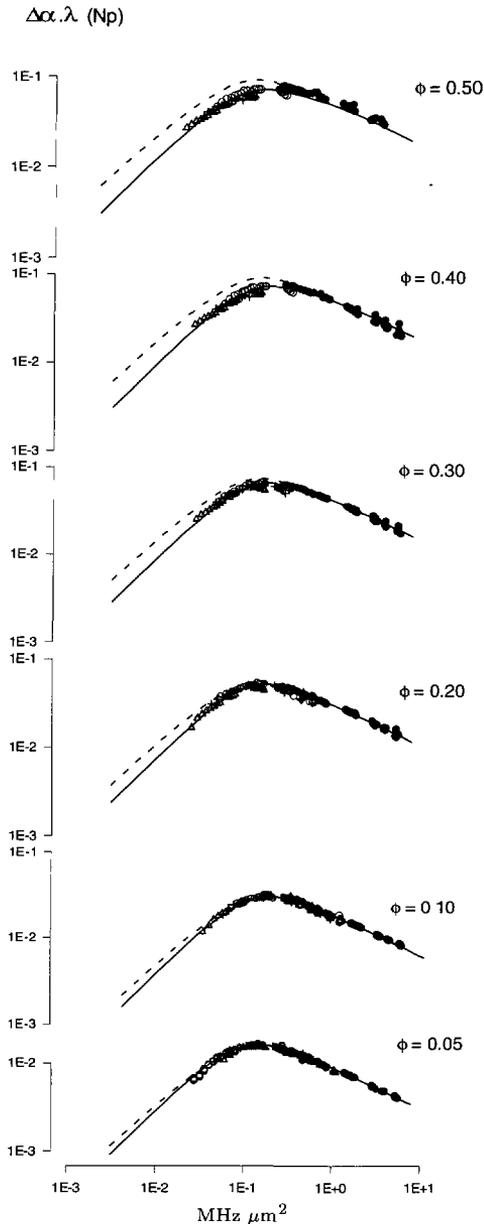


Fig. 3

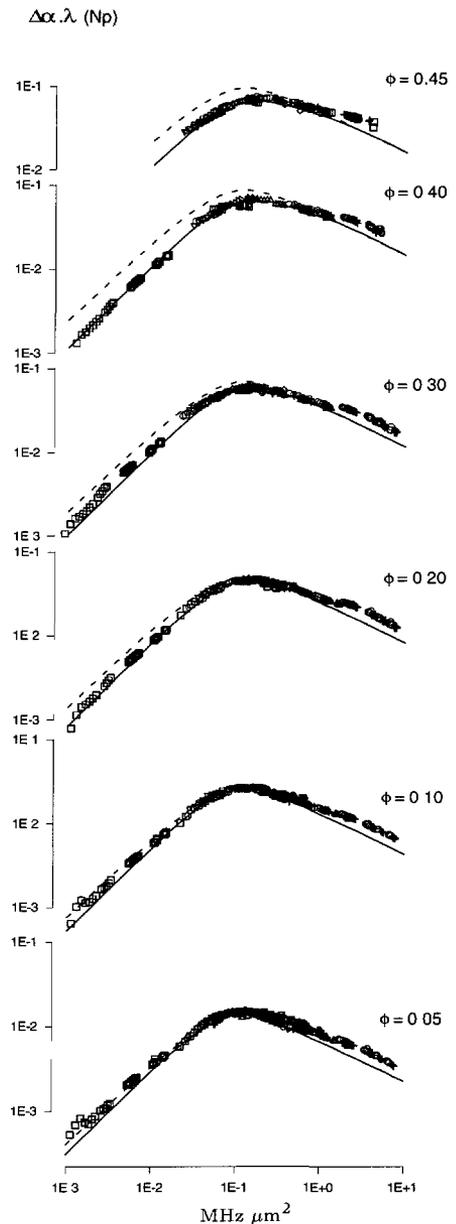


Fig. 4

Fig. 3. — Comparison between the core-shell model (solid lines) and the model of Isakovitch (dashed lines) for 4 monodisperse emulsions of polysiloxane oil in water, the sizes of the droplets range between 230 nm and 760 nm, the volume fraction varies from 5% to 50%. For each sample the value of the radius has first been determined by a least-squares fit of the model of Isakovitch to the spectrum measured at 10% volume fraction. The values thus obtained are then used to place each spectrum, at each concentration, on the  $f \cdot a^2$  plot.

Fig. 4. — Comparison between the core-shell model (solid lines) and the model of Isakovitch (dashed lines) for 10 hexadecane/water emulsions, the sizes of the droplets range between 46 nm and 900 nm, the volume fraction varies from 5 to 45%. The deviation visible at high frequency is due to the visco-inertial effect induced by the slight difference between the densities of n-hexadecane and water [8].

Table I. — *Thermo-physical constants of materials at 25 °C (taken from Ref. [8]).*

	n-hexadecane	silicone oil	aqueous phase
Sound velocity $c$ (m s <sup>-1</sup> )	1357.9	1004	1482
Density $\rho$ (kg m <sup>-3</sup> )	773.0	975	998.2
Thermal expansivity $\beta$ (K <sup>-1</sup> )	$9.1 \times 10^{-4}$	$9.4 \times 10^{-4}$	$2.13 \times 10^{-4}$
Specific heat $C_p$ (J kg <sup>-1</sup> K <sup>-1</sup> )	2215	1460	4182
Thermal conductivity $\kappa$ (W m <sup>-1</sup> K <sup>-1</sup> )	0.143	0.15	0.591

size based on this model would lead to underestimating the radius by more than 25%.

In the case of hexadecane emulsions a divergence from the model can be noticed at high frequencies. This difference is to be assigned, as shown in reference [8], to the visco-inertial effect arising from the non-negligible density difference between water and hexadecane. In the case of polysiloxane emulsions the fact that the densities of the two media are nearly equal explains why the model matches so accurately the experimental results, in this case the thermo-elastic effect is the dominant loss mechanism. The price to pay is that the preparation of small droplets is much more difficult, so that the spectra are limited to higher values of the variable  $f \cdot a^2$ .

#### 4. Discussion, Conclusion

The above results establish that the core-shell model offers a very accurate prediction of the ultrasonic attenuation in concentrated emulsions, up to 50% in oil-volume fraction, without resorting to any adjustable parameter. For applying the model in actual practice it is important however to be aware of the conditions in which it is pertinent. Namely the other sources of attenuation must be either negligible or known with enough accuracy. Two mechanisms may be responsible for such losses: the visco-inertial effect and the scattering of the elastic wave. Existing theories are able to describe these effects accurately when the concentration is low enough; however for higher concentrations the modelization of multiple scattering, on one hand, becomes increasingly difficult, and we are not aware of any theory taking into account the hydrodynamic interactions between particles which may affect the visco-inertial effect on the other hand.

Scattering losses may be safely predicted to be negligible, for liquid-liquid suspensions, if the frequency range does not exceed 10 MHz and if droplets sizes remain under a few microns. As regards the visco-inertial losses, the results that we have obtained on hexadecane emulsions indicate that, despite a significant density contrast, their influence does not affect seriously the determination of the size of droplets in the low  $f \cdot a^2$  regime.

It must also be observed that our model stands for monodisperse systems, we do not know how to manage the case of polydisperse systems. It seems impossible to be satisfied with a mere superposition of the contributions of all size classes, as particles of different sizes will interact with each other. Measurements performed on mixtures of known fractions would be useful in this respect.

The model predicts that the losses must vanish when the volume fraction tends toward unity, which is obviously consistent because there would be no heat exchanges in pure oil. Measurements on highly concentrated creams would thus be interesting, in order to determine up to which concentration the predictions remain valid. Finally we think that a reason why our

model is successful is due to the fact that it is adequate at both ends of the frequency domain. At high frequencies it reduces to the model of Isakovitch which is fundamentally correct because each droplet acts separately. At low frequencies, as the particle interacts with more and more neighbor particles, and because thermal equilibrium between each droplet and the surrounding water can be reached within the period of the wave, then the idea of an effective medium appears to be adequate. As the ultrasonic absorption varies in a very monotonous way when the frequency increases, the connection between the two regimes cannot deviate much from the reality.

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