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Running title

Ultrasonic waves in heterogeneous solids

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Abstract

This study compares ultrasonic wave propagation modeling and experimental data in concrete. As a consequence of its composition and manufacturing process, this material has a high elastic scattering (sand and aggregates), and air (microcracks and porosities) content. The behavior of the "Waterman-Truell" and "Generalized Self Consistent Method" dynamic homogenization models are analyzed in the context of an application for strongly heterogeneous solid materials, in which the scatterers are of various concentrations and types. The experimental validations of results predicted by the models are carried out by making use of the phase velocity and the attenuation of longitudinal waves, as measured by an immersed transmission setup. The test specimen material has a cement-like matrix containing spherical inclusions of air or glass, with radius close to the ultrasonic wavelength. The models are adapted to the case of materials presenting several types of scattering particle, and allow the propagation of longitudinal waves to be described at the scale of materials such as concrete. The validity limits for frequency and for particle volume ratio, can be approached through the comparison with experimental data. The potential of these homogenization models, for the prediction of phase velocity and attenuation in strongly heterogeneous solids, is demonstrated.

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I. INTRODUCTION

The general context of this study is the evaluation of damage in concrete structures using ultrasonic methods. Damage often leads to the appearance and then to the development of microcracks and/or microporosities in the medium^{1,2}. This type of damaged medium can be described as a heterogeneous medium composed of an elastic matrix (the cement) containing elastic inclusions (the aggregates) and air inclusions (the microcracks). Interaction between the wave and the obstacles leads to scattering of the wave. In the case of concrete, in which there are numerous obstacles, it is necessary to take multiple scattering effects into account^{3,4}.

The observable parameters, which are the velocity and attenuation of ultrasonic waves, are difficult to relate to the size of the aggregates, the volume fraction of porosities, the elastic constants, and damage in the medium. The understanding of the direct problem in concrete requires the propagation phenomena of ultrasonic waves, in heterogeneous media containing high densities of different types and shapes of scattering particle, to be modeled. The experimental validation phase of the models not only demonstrates the potential and limitations of this type of model, but also highlights the work which still remains to be accomplished^{3,5,6}.

The prediction of propagation in a multi-scattering particle medium can be made using the exact calculation of the field scattered by all of the obstacles. However, the large number of scattering particles and the need to know the position, type, size and shape of each of them rapidly lead this technique to become costly in terms of computation time, and very difficult to implement whenever some of the parameters are random, such as the exact positions and shapes of the scatterers. Medium homogenization methods, which integrate the multiple scattering equations, can thus be a good modeling solution for a wave propagating through an acoustically equivalent homogeneous medium. There are numerous medium homogenization models. The simplest ones are independent of frequency, take only simple scattering into account, and are of the static or quasi static type: Kuster and Toksöz⁷, Berryman⁸ for example. These models are limited to use in the Rayleigh domain, in which the wavelength is much greater than the size of the obstacle, and for scattering particle concentrations which are often low.

If the size of the inclusions is of the order of one wavelength, the wave propagation characteristics such as phase velocity and attenuation become frequency dependent³⁻⁶. In the stochastic domain, many dynamic models provide a good description of propagation. The integration of multiple scattering effects combined with the application of a statistical mean to the positions of isotropic scattering particles was initiated by Foldy⁹. The extension of this model to non-isotropic scattering particles was proposed by Waterman and Truell¹⁰ and Twersky¹¹, and modified versions of these methods were proposed more recently by Linton and Martin¹². In parallel with these developments, hypotheses concerning the relative positions of the scattering particles were formulated by Lax¹³, who proposed the quasi-crystalline approximation (QCA). This approximation was coupled to different correlation functions, which led to various models such as those of Tsang¹⁴, Varadan¹⁵, and Sabina and Willis¹⁶. For these models, the choice of inter-correlation functions is important, and solutions are difficult to obtain in the short wavelength domain. The numerical computations thus become complex and their convergence towards a solution is no longer certain.

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Finally, other methods rely on one or another of the two preceding families, but use an iterative process which makes it possible to converge towards the wavenumber of the equivalent medium: Kim¹⁷ proposed an initial auto-coherent scenario, which he called the Coherent Potential Approximation (CPA). Another approach based on the same principle was proposed by Yang¹⁸: the Generalized Self Consistent Model (GSCM). Its results are close to those of Waterman and Truell's model¹⁰ and to the experimental comparisons with lead/epoxy composites carried out by Kinra and Rousseau¹⁹. These auto-coherent methods are described as having potentially good performance in strongly heterogeneous media.

In the context of an application in solid media with high scattering particle concentrations, this study is based on the Waterman and Truell model (WT), the formalism of which is by far the simplest, and which has already demonstrated promising results³, and the Generalized Self Consistent Model (GSCM) proposed by Yang which has a strong potential for the study of highly charged media.

More generally, the experimental validations related to these models are numerous in the case of mediums with a fluid matrix, but more rare in the case of solid matrices. In this case, the models' limitations, in terms of the frequency range or the volume fraction of the obstacles in solids, need to be defined.

The application of these models to the case of concrete is envisaged. As a consequence of the shape and the nature of the obstacles, the problem is analyzed for three dimensions objects over a wide range of frequencies, in order to cover the potential domain of ultrasonic testing applications. The cases of elastic inclusions, as well as that of air inclusions and the combination of both types of inclusions, are studied. This paper proposes to compare the modeling results of the two effective theories with experimental data

obtained in concrete media. The long-term objective is to be able to predict the acoustic behavior of waves in damaged concrete.

In Section II, we describe the models we used, their extension, allowing several types of inclusions to be accounted for, and their behavior with respect to the media analyzed in the present study. The experimental study, described in Section III, presents the test specimens, the acquisition protocol and the measurement principle. In Section IV, we compare the results provided by the models and the experiments. Finally, our conclusions are given in Section V.

II. DYNAMIC HOMOGENIZATION MODEL

A. Homogenization principle

In order to compute the field scattered by the full set of obstacles, the theory of equivalent media, or homogenization, allows a heterogeneous medium to be replaced by an acoustically equivalent homogeneous medium (Fig. 1).

These propagation models are based on the statistical average of multiple scattering equations, relying on the acoustic scattering behavior of a single object. Spherical geometries are considered in this study, because of their similarity to the shape of real objects present in concrete, such as aggregates or porosities. Although the microcracks are less similar to spheres, nevertheless we assume the hypothesis that the scattering effects arising from a set of randomly oriented microcracks can have similar behaviours than a set of randomly distributed spheres. So we describe the scatterers by a distribution of spheres of rocks or air. The propagation characteristics of the longitudinal wave, i.e. the phase velocity c^* and the attenuation α^* are respectively the real and imaginary parts of the complex wavenumber k^* (the asterisk indicates the equivalent medium):

$$k^* = \frac{\omega}{c^*} + i.\alpha^*$$
(1)

where $\omega = 2.\pi$.f is an angular velocity and f is the frequency.

This wavenumber for the equivalent homogeneous medium, is this one defined using the dynamic homogenization models, and depends on the wavenumber in the matrix, the ones in the inclusions, and the sizes and densities of the inclusions.

B. The Waterman-Truell model

In the initial approach (Fig. 2) the scattering particles are all identical, with a radius a, a volume fraction inside the medium which is assumed to be constant and equal to τ , and positions which are statistically independent. In the case of spherical scattering particles, the results for a longitudinal wave propagating in the equivalent medium lead to the following relationship for the complex wavenumber¹⁰:

$$\left(\frac{k^{*}}{k_{1}}\right)^{2} = \left[1 + \frac{2.\pi .n_{0}.f(0)}{k_{1}^{2}}\right]^{2} - \left[\frac{2.\pi .n_{0}.f(\pi)}{k_{1}^{2}}\right]^{2}$$
(2)

where $n_0 = \frac{3.\tau}{4.\pi a^3}$ is the number of scattering particles per unit volume, K_1 is the

wavenumber in the matrix and f(0) and $f(\pi)$ are the forward and backward form function for a single scattering particle. The scattering properties of a single object are characterized by the form function. This gives the angular distribution of the amplitude of the scattered wave surrounding the obstacle, in particular in the forward and backward directions. In the case of longitudinal waves the form function, for an observation angle θ is given by the following expression²⁰:

$$f(\theta) = \frac{1}{ik_1} \cdot \sum_{n=0}^{\infty} (2.n+1) \cdot (T^{11})_{n0n0} \cdot P_n^0(\cos(\theta))$$
(3)

This far-field expression is based on the decomposition of potentials using the Legendre polynomials (P_n^0), and on the use of the T-matrix $(\overline{T})^{20}$ in the scattering calculations. $(T^{11})_{n0n0}$ is the coefficient, of n order, derived from this matrix, corresponding to the longitudinal wave scattering of a longitudinal wave. The expression for this coefficient in the case of a spherical, fluid or elastic object in an elastic medium is computed from the ultrasound parameters of the matrix and the scattering particles using a resonant scattering theory²¹. This technique is widely used in the scattering calculations and had been experimentally validated for the case of elastic sphere in elastic medium²² with very good agreement.

C. The "Generalized Self Consistent Method" model

Yang¹⁸ considered a scattering medium composed of an equivalent matrix in which scattering particles are included (Fig. 3). As opposed to the previous model, the equivalent matrix corresponds to a homogenized medium instead of the real matrix. This is the hypothesis which makes the model self-coherent. It is an iterative approach which converges, tending towards the characteristics of the equivalent medium.

The scattering particles are all identical, with a radius a, and have a volume fraction in the medium assumed to be constant and equal to τ , and statistically independent positions. These scattering articles are thus considered to be inside a shell of characteristic size b having the same acoustic properties as the real matrix.

In the case of spherical diffusers, the radius b of the shell is related to the radius a of the obstacle by the obstacle volume fraction in the medium, written as:

$$\tau = \frac{a^3}{b^3} \tag{4}$$

The result of the "Generalized Self Consistent Method" is obtained from equation (2), with the wavenumber of the matrix k_1 being replaced by that of the homogenized medium k*. For the longitudinal wave propagating in the equivalent medium, we thus obtain the following expression:

$$1 = \left[1 + \frac{2.\pi . n_0.f_{gscm}(0)}{k^{*2}}\right]^2 - \left[\frac{2.\pi . n_0.f_{gscm}(\pi)}{k^{*2}}\right]^2$$
(5)

where $f_{gscm}(0) = \sum_{n=0}^{\infty} (-i)^n . A_{0n}$ and $f_{gscm}(\pi) = \sum_{n=0}^{\infty} (i)^n . A_{0n}$ (6)

These are the forward and backward scattering functions of a longitudinal wave impinging on a spherical obstacle of radius a, included inside a shell of radius b, which itself is embedded in an equivalent matrix. These functions are calculated starting from the scattering coefficient A_{0n} defined by Yang¹⁸. Taking into account inclusions inside a shell of radius b (eq. 4) depending on volume fraction of inclusion induces modifications of scattering functions. This way to model inclusions in 2D effective medium model coupled with the self-consistent scheme provided good agreements with experiment in the SiC/Ti fiber/matrix composite²³ and in particular at high concentration.

Although equation (5) cannot be solved directly, an iterative pattern initialized by the result from equation (3) can be used to find the solution. The iterative pattern is then stopped when the results converge.

D. Taking different obstacles into account

In the case where there are several sizes and types of scattering particle, it is possible, for both models, to introduce (in equations 3 and 6) the influence of these scattering particles, based on the average of the scattering functions, such that:

$$\langle f(\theta) \rangle = \sum_{i} \frac{n(i)}{n_0} f(\theta, i)$$
 (7)

where i is the index of the different scattering particles, n(i) is the number of particles (i) per unit volume, and $f(\theta,i)$ is the scattering function of the obstacle i, at an angle θ .

E. Modeled outcome for wave propagation in concretes

In order to observe the evolution dynamics of the two propagation models, the responses of the models are studied, in terms of phase velocity and attenuation, as a function of the longitudinal wave's frequency in media similar to concrete. The scattering particles related to the composition and to the damage are introduced separately, then simultaneously. The maximum order used in scattering calculations (eq. 3 and 6) for n is 20, at this value we didn't observed significant variations in function forms for the studied frequency domain. Variations in the volume fraction of the scattering particle are proposed.

The model is based on the knowledge of a mortar matrix ($\rho = 2145 \text{ kg/m}^3$) in which spherical scattering particles have been introduced. The longitudinal ($c_L = 4387 \text{ m/s}$) and transverse ($c_L = 2492 \text{ m/s}$) propagation velocity characteristics of the matrix are constant. A second order polynomial approximation describes the attenuation as a function of frequency (Fig. 4). The measurement protocol used to obtain these values is that shown in Section III.B.

The variation of the attenuation is approximated by a second order polynomial function, which corresponds to the best approximation found for all orders between 1 and 4. Although this approximation is valid for the domains corresponding to the passband of the sensors (from 200 kHz to 1.2 MHz), the classical range of test frequencies rarely exceeds the value of approximately 500 kHz, which is included by the sensor passband.

Table 1 presents the materials used for the scattering particles into the mortar matrix. Glass density was provided by glass beads manufacturer and glass ultrasonic velocities were chosen near to literature values²⁴. The value for the longitudinal velocity has been verified, on several beads of 10 mm diameters, by contact transmission measurement. Expanded polystyrene parameters included in specimens were considered equal to the ones of air according to the manufacturer which ensures that the material contains 98 percent air.

In order to tend towards the real composition of concretes, glass beads were introduced into the mortar matrix. Glass was chosen for its physical, mechanical and acoustic properties which are close to those of the rocks normally found in concrete. The simulation of damage in concrete is implemented by introducing spherical cavities. This damage often occurs through the appearance or evolution of porosities or microcracks in the mortar matrix.

1. Modeling concrete composition

The numerical application relates to the two studied models, which we note as WT for the Waterman-Truell's model¹⁰, and GSCM for that of the "Generalized Self Consistent Method"¹⁸. The introduced media are identical for both models, and correspond to 10%, then 30% of 3 mm diameter spherical glass diffusers in the fine mortar matrix. The maximum chosen values of volume fraction correspond to values similar to those encountered in concrete, for a given obstacle dimension. The dimensions of aggregates in media such as concrete can vary between 0 and 20 mm; the simulation relates to beads with a diameter of 3 mm. The results obtained in terms of phase velocity and attenuation as a function of frequency are shown in Fig. 5.

The introduction of solid inclusions whose acoustic impedance is greater than that of the matrix leads, whatever the model, to an increase in phase velocity and attenuation relative to those obtained for the matrix alone (Fig. 4). The increase in velocity results from the higher velocity in the inclusions, and the increase in attenuation results from the scattering produced by the introduced objects. The greater the volume fraction of scattering objects, the greater the velocity and attenuation values over the studied frequency domain.

We observe different behaviors for the two models, in terms of their dependence on frequency. When the scattering particle volume fraction increases, the differences between the two models become stronger. In terms of velocity analysis, we observe, for both models, a velocity minimum at a low frequency (around 450 to 500 kHz, which corresponds to values of k.a close to 1) for 10% of scattering particles, and a slightly higher value (nearer to 500 kHz, with k.a close to 1) for 30% of scattering particles.

The evolution of the attenuation also reveals different behaviors for the two models, between which the greatest differences are observed at high frequencies. Over the first part of the frequency domain (< 500 kHz, k.a < 1), the models are relatively close to one another. Strong differences are observed between the two models at higher frequencies, with diverging values of attenuation. More the volume fraction of glass inclusions are high, more the observed differences are important. These behaviors confirm those observed by Yang¹⁸ in glass/epoxy composites.

2. Modeling damage in concretes

In order to model damage in a medium, the 3 mm diameter spherical cavities are introduced into the same fine mortar matrix, with volume densities of 10%, and then 30%. The value of 30% of air inclusions in a granular medium is high in comparison with the evolution in volume expected when such media are damaged. The results obtained in terms of phase velocity and attenuation, as a function of frequency, are shown in Fig. 6.

Whatever model is used, the introduction of air inclusions with an acoustic impedance lower than that of the matrix leads to a fall in velocity and an increase in attenuation, in comparison with the values obtained for the matrix alone (Fig. 4). The fall in velocity is caused by the slower velocity in the inclusions, and the increase in attenuation is caused by the scattering produced by the introduced obstacles.

The greater the volume fraction of the scattering objects, the greater the changes in velocity and attenuation over the full domain of studied frequencies.

Similar curves are obtained for both models. However, the differences between the models increase with increasing scattering particle volume fraction.

We systematically observe a fall in velocity at low frequencies, with a local minimum in the vicinity of 450 to 500 kHz, for 10% of scattering particles (k.a close to 1) and slightly greater (closer to 500 kHz, k.a close to 1), for 30% of scattering particles. The strongest differences between the WT and GSCM models are observed at low frequencies. For the attenuation, we also note similar behaviors, with transitions (sudden changes in slope) occurring at different frequencies. Like for the case of composition modeling, one can observe increasing differences between the two models with increasing scatterers volume fraction.

3. Coupling between composition and damage

The coupling of two types of scattering particle is produced by simultaneously introducing 10% of spherical glass scattering particles with a diameter of 3 mm, and 10% of spherical cavities with a diameter of 3 mm, into a fine mortar matrix. This operation allows the coherency of the results given by this simulation, in comparison with the results of the preceding simulations, and the models' respective abilities to simultaneously integrate several types of diffuser, to be evaluated. The results obtained in terms of phase velocity and attenuation as a function of frequency are shown in Fig. 7.

The results obtained clearly show the combined effects of the different separately observed behaviors. The differences between the models are less significant than in the two preceding cases, which can be explained by the behavior inversions depending on the model, with respect to the different types of scattering particle. As the glass and cavities in this simulation identical in size, the strong evolutions obtained at 500 kHz (k.a close to 1 in both cases) are quite noticeable for both models. The velocities are distributed around that found for the matrix alone. The greatest divergences between the two models are observed in the high frequency domain.

The variations in simulated attenuation are very similar, which can be explained by the inversion of the attenuation behaviors between the models, depending on the types of scattering particle. The attenuation values are of course greater than when each case is treated separately.

III. EXPERIMENTAL VALIDATION STUDY

The experiments we carried out were designed to verify the validity of the wave propagation models, and to evaluate the limits of each model with respect to the types, sizes and densities of scattering particles, as well as in terms of the frequency of the acoustic waves used for the tests.

A. Definition of the test specimens

All of the test specimens we used were made from the same, very fine mortar matrix obtained with a mixture of cement, water and fine sand (grain size < 0.5 mm). This matrix is defined in Table 2, and is referred to as FM (Fine Mortar). Glass beads of different sizes (diameter \emptyset) were then added to the FM in order to simulate the composition of concretes (FM-G3 and FM-G10), and expanded polystyrene balls (98% air) allowed damage to be simulated by increasing the air scattering volume fraction inside the medium (FM-P3). The

3 mm beads allowed sand grains to be accounted for, and the 10 mm beads were used to represent aggregates. The air beads can simulate damage by increasing the porosity of concretes and/or damage caused by the appearance and evolution of randomly oriented microcracks. The coupling between composition and damage is achieved by using a test specimen containing both types (glass and air) of diffuser (FM-G3-P3).

The fine mortar test specimen (FM) allows the characteristics of the matrix, into which the scattering particles are later introduced, to be determined. The matrix starting values for the models are those measured and presented in Fig. 4. The data concerning the glass beads and expanded polystyrene balls included into the test specimens are shown in Table 1. The expanded polystyrene balls are composed of 98% air, and are considered as equivalent to air inclusions. The test specimens have the shape of a disk, with a diameter of 250 mm and thickness of 50 mm.

B. Experimental measurement protocol

The ultrasound measurement protocol we developed was adapted to the characterization of heterogeneous media. The measurements were carried out using the immersion transmission of a longitudinal wave, which allowed reproducible coupling levels to be obtained between the specimens and the transducers. Three pairs of transducers were used and the total passband found at -12 dB ranged from 200 kHz to 1.2 MHz. The measurement was based on a comparative method between the direct signal transmitted through water and that corresponding to a trajectory through water and the tested specimen (Fig. 8).

The distance between the sensors was the same in both cases, with the specimen being placed in the far field of the emitting transducer. The quantities averaged over the scatterers positions are obtained from averages, for each specimen, on uncorrelated transducers positions. This operation allows the amplitude of the incoherent waves to be reduced, thus allowing the coherent transmitted wave to be separated and analyzed. The broad-band temporal signals obtained are reduced by Fourier analysis, in order to extract their phase and amplitude.

The phase velocity and attenuation of the ultrasonic sensor are then determined, from the thickness of the specimen (e) and the following expressions:

$$c_{exp} = \frac{\omega e}{\phi_{s}(\omega) - \phi_{w}(\omega) + \frac{\omega e}{c_{w}} + \Delta \phi_{D}(\omega)}$$
(8)

$$\alpha_{exp} = -\frac{1}{e} . \ln \left[\frac{D(d, \omega)}{T_{w/s}(\omega) T_{s/w}(\omega) D(d, e, \omega)} \cdot \frac{S_s(\omega)}{S_w(\omega)} \right]$$
(9)

The terms $\phi_w(\omega)$ and $S_w(\omega)$, as well as $\phi_s(\omega)$, are respectively the phases and amplitudes of the signals obtained through water, and those obtained through water and the sample. The coefficients $T_{w/s}(\omega)$ and $T_{s/w}(\omega)$ are the transmission coefficients at the water-specimen and specimen-water interfaces respectively. The terms $\Delta \phi_D(\omega)$, $D(d,\omega)$ and $D(d,e,\omega)$ are the corrective coefficients for phase and amplitude, respectively, of the beam divergence in the far field during transmission of the acoustic waves. These terms are computed from the Thompson model²⁵. The measurement uncertainties were evaluated for the full bandwidth, using repeatability tests. This uncertainty varies only slightly as a function of frequency and the type of test specimen. The values retained were the maxima, i.e. ± 45 m/s for the phase velocities and ± 3 Np/m (values corresponding to a 95.4 percent confidence interval). These values are given on the following figures by dotted curves.

IV. RESULTS AND DISCUSSION

In order to validate the behavior of the studied models during the previous numerical simulations, the present section proposes a comparison between theory and experiment, based on the specimen compositions described in Table 2. Firstly, the results for test specimens with compositions simulating similar concrete formulations (FM-G3 and FM-G10) were studied. Secondly, the results for test specimens simulating damaged concrete (FM-P3), and thirdly results from coupling between two types of scattering particle, based on the (FM-G3 P3) test specimen, are presented.

Fig. 9 shows the velocity and attenuation results found for the FM-G3 test specimen made of fine mortar including 34.1% of 3 mm diameter glass beads, with the corresponding models.

The phase velocity results are found to be in nearly perfect agreement between the experiment and the GSCM model, in the observed experimental passband. The maximum differences between the two curves remain within the measurement uncertainties (dotted curves). Dispersion on measured phase velocity at low frequency can be attributed to electrical noise that can occurred for small signal to noise ratio²⁶ especially at the limit of the passband.

As far as the attenuation is concerned, the general shape of the curves is close to that of the WT model, although the experimental values obtained are quite different to those given by

this model. They are close to the GSCM model values in the low frequency part of the passband, and are removed from them at frequencies greater than 700 or 800 kHz (k.a is approximately equal to 1.5). The overestimating behavior of the WT model is classically observed in literature for acrylic spheres in ethylene glycol²⁷ and for glass spheres in epoxy matrix²⁸. The good agreement of the GSCM model for high frequencies is also obtained the solid heterogeneous medium²⁸.

Fig. 10 shows the velocity and attenuation results relative to the FM-G10 test specimen composed of fine mortar including 32.1% of 10 mm diameter glass beads, with the corresponding modeled curves. This allows differing behaviors to be observed, as a function of changes in size, and possible limits in useful range of the models, in terms of frequency or scattering domain, to be observed.

The results for phase velocity show that there is a good agreement between the experimental and GSCM model values, between 200 kHz and 700 kHz (k.a is approximately 5), the latter frequency being that beyond which the measured velocity begins to fall. This decrease in velocity then brings the experimental behavior closer to that predicted by the WT model at high frequencies. Theses behaviors can be related to the one observed by Layman and al^{28} in case of glass spheres in epoxy matrix in which no conclusion can be done between WT and GSCM models for the phase velocity dispersion. The obtained results for the velocity in the present study show better agreements for GSCM model than WT one when elastic spheres are introduced in elastic matrix.

Concerning the attenuation, the general shape of the curves is close to that of the GSCM model and the values obtained are generally very close to those given by the model. As previously shown (for 3 mm beads), the experimental values depart from the model at

frequencies higher than 400 kHz (k.a values approximately equal to 2.5). The greatest differences observed are 10 Np/m at the highest frequencies (1.2 MHz).

The GSCM model thus appears to more accurately describe the behavior of ultrasonic waves, when the scatterers introduced into the elastic matrices are elastic spherical particles.

In order to evaluate the influence of introducing cavities into the medium, Fig. 11 shows the velocity and attenuation results for the FM-P3 test specimen. It is made of fine mortar, with 30% of 2.84 mm diameter expanded polystyrene balls, together with the results of the corresponding model.

The phase velocity results show good agreement between the experiment and the WT model, over the full frequency domain. The differences between experiment and WT model remain very near to the measurement uncertainties limits (dotted curves). The two local minima predicted by the GSCM model do not appear in the experiment, for which just one minimum can be observed.

The attenuation curves show that the measured values are very close to the WT model, up until 700 kHz (k.a = 1.6), and then become greater than those of the model beyond this frequency. The general shape of the experimental and WT modeled curves are very similar. The attenuation foreseen by the GSCM model is quite different to the measured values. This behavior should be complemented by Fig. 6, which shows that beyond the observed frequencies the curve should continue to increase, up until a turning point at a much higher value. Measured attenuation at high frequency reaches important values and has a dispersive behavior that can be related to the one obtained at low frequency on velocity on Fig. 9.

The WT model appears to have the best performance in describing the behavior of ultrasonic waves when spherical cavities are introduced into elastic matrices.

Finally, Fig. 12 presents the phase velocity and attenuation results corresponding to the FM-G3-P3 test specimen combining two types of diffuser. This specimen comprises fine mortar including 34.1% of 3 mm diameter glass beads and 17.1% of 2.84 mm diameter expanded polystyrene balls.

The results for phase velocity show that there is a good agreement between the shape of the experimental and studied model curves, over the full frequency range. However, there are significant differences between the theory and the models, in particular for the WT model. The GSCM model is in agreement with the experimental values beyond 700 to 800 kHz (k.a is of the order of 1.5). These observations should be compared with the velocity curves shown in Figs. 9 and 11, where it can be seen that the WT model does not fully represent the solid scattering particles, and the GSCM has evolution dynamics which are too strong (in the vicinity of k.a values close to 1), in the case of the introduction of the cavities.

The attenuation computed by the GSCM model is close to the experimental values. The WT model predicts an attenuation curve similar in shape to that found experimentally, but with higher values than in the experiment. Neither of these two models allows a quantitatively correct description to be found, in agreement with the experimental measurements. Their good qualitative agreement nevertheless confirms the potential of these models in this type of medium.

The different results show the good agreements of GSCM model for the case of elastic inclusions in elastic matrix and good agreements of WT model for the case of

cavities in elastic matrix up to about k.a=1.5 and when volume fraction of particles reaches 34%. The frequency limit is sufficient to cover the typical frequency range of ultrasonic testing methods for concrete and the volume fractions of obstacle are representatives of the ones obtained in concrete material.

Combination of the two types of scatterers provides relative agreements but not quantitatively perfect in particular for the low frequencies range. This case highlights the potential of effective medium modeling to reach high volume fraction but also improvements we need to do. The observed differences in results between model and experiment can be due to different sources.

First, small errors in measurement at the limits of the total passband²⁵ (see Fig. 9 for velocities at low frequencies or Fig. 11 for attenuation at high frequencies) can appear on measured velocities and attenuations. Another parameter is the averaging process on scatterers positions done by uncorrelated spatial averaging which can be quite different or particular because of the nature of statistic observations. Second, all parameters used for the input data (Tables 1 and 2) are known with unknown uncertainties and may be inaccurate due to difficulties to obtain representative values in literature (properties of glass and expanded polystyrene), to control manufacture process (real values of volume fractions of inclusions, similar cement matrix for all specimen, ...) or to implement good evaluation of this parameters by measurement systems. Third, approximations done in the construction of the different models can be definitely the largest part of the observed disagreements. To comment this point we can focuses on each case of inclusion type (see Fig 9 and Fig. 11). In the two models, differences can appear due to mismatch of approximation in calculation for the scattering by one obstacle or approximation in the construction scheme of effective

medium model. If the way to model scattering by sphere used in WT model is classically employed and is validated with good accuracy, the one used in GSCM model, with transition medium around the sphere, is quite different and cannot be validated separately. This method is coupled with the self-consistent scheme and shows, in past study²³, interesting improvement for the elastic two dimensional case (scattering by elastic cylinders). For the case of elastic three dimensional spheres, we also observed improvement of results provided by GSCM model in comparison to WT model. This is not true for the case of cavities in elastic matrix. The way used in GSCM model seems to be adapted for elastic inclusions and to be failed for cavities. Maybe the strong acoustic contrast between inclusions and matrix induces too high variations in scattering calculations done in GSCM model.

The two employed models are originally constructed on the basis of fluid media including scatterers with existence of an only one longitudinal wave in the effective medium and the conversion modes are neglected in this multiple scattering theory²⁹. So, the energy involved in mode conversions is considered as lost that can induces overestimating of attenuation and also dispersion in phase velocity. If the self-consistent scheme provides new solutions in case of high concentrated elastic scatterers in elastics matrix, taking into account mode conversions using Quasi Crystalline Approximation^{13,12,29} will surely improve modeling results, like it is already done for two dimensional random composites³⁰.

V. CONCLUSION

We have analyzed the longitudinal wave propagation in a heterogeneous medium like concrete including elastic or fluid scattering particles. The evolution of the phase velocity and the attenuation have been computed with two homogenization models (Waterman and Truell - WT, and "Generalized Self Consistent Method" - GSCM) over a wide range of frequencies, as a function of the volume ratio and size of spherical scattering particles. An experimental validation study has allowed each of these models to be qualified with respect to changes in concrete. Both models have shown different potential, with the GSCM appearing to be the most suitable for describing elastic solids containing elastic scattering particles, whereas the WT model allows cavities to be taken into account. The observed frequency limits are found to be in good agreement, up until k.a values of the minimum order of 1.5 for the velocity and for the attenuation. Both models have shown good potential in representing the two types of scatterers. The key to modeling improvements may lie in the coupling of these two models, by way of an incremental approach to their representation for different types of scattering particles. A first step can be the modeling of concrete composition by elastic inclusions in a cement matrix with GSCM model to obtain equivalent homogeneous medium. Then this equivalent medium can be used like matrix for the second step including spherical cavities with WT model to take into account damage in concrete. Thus, with these two steps, one can reach high volume fraction of obstacles in elastic matrix.

This outcome allows the use of these models to be validated in media with very high scattering particles volume fraction, for the prediction of phase velocity and attenuation but neglecting multiple scattering effects between the two types of scatterers. Another way to improve ultrasonic non destructive characterization of heterogeneous media with high particles concentration can be conducted using model which integrates modes conversion.

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Tables

TABLE 1. Properties of the media under consideration

Media	ρ (kg/m ³)	c _L (m/s)	c _T (m/s)
Glass	2650	5700	3200
Air	1	330	-

Ref.	Cement (%)	Water (%)	Additive (%)	Fine Sand Ø<0.5mm (%)	Glass beads Ø3mm (%)	Glass beads Ø10mm (%)	Polystyrene Balls Ø 2.84mm (%)
FM	28.5	39.7	0.9	30.9	-	-	-
FM-G3	18.7	26.2	0.7	20.3	34.1	-	-
FM-G10	19.2	26.8	0.6	21	-	32.1	-
FM-P3	19.9	27.9	0.6	21.7	-	-	30
FM-G3-P3	13.8	19.5	0.4	15.1	34.1	-	17.1

TABLE 2. Test specimen compositions

Figure captions

FIG. 1. Real medium, geometric model and equivalent homogeneous medium.

FIG. 2. Homogenization in the Waterman-Truell model.

FIG. 3. Homogenization in the "Generalized Self Consistent Method" model.

FIG. 4. (Color online) Velocity and attenuation versus frequency of the longitudinal wave (experiment points with approximation - dashed lines) in a fine mortar matrix (FM).

FIG. 5. (Color online) Phase velocity and attenuation modeled versus frequency in fine mortar containing 10% (continuous lines), and 30% (dashed lines), of 3 mm diameter glass beads.

FIG. 6. (Color online) Phase velocity and attenuation modeled versus frequency in fine mortar containing 10% (continuous lines), and 30% (dashed lines), of 3 mm diameter spheres of air.

FIG. 7. (Color online) Phase velocity and attenuation modeled versus frequency in fine mortar containing 10% of 3 mm diameter glass beads and 10% of 3 mm diameter spheres of air; WT model (continuous lines) and GSCM (mixed lines).

FIG. 8. Comparative measurements in immersion.

FIG. 9. (Color online) Modeled (WT: continuous lines, and GSCM: mixed lines) and experimental (points) phase velocity and attenuation versus frequency in fine mortar containing 34.1% of 3 mm diameter glass beads (FM-G3).

FIG. 10. (Color online) Modeled (WT: continuous lines, and GSCM: mixed lines) and experimental (points) phase velocity and attenuation versus frequency in fine mortar containing 32.1% of 10 mm diameter glass beads (FM-G10).

FIG. 11. (Color online) Modeled (WT: continuous lines, and GSCM: mixed lines) and experimental (points) phase velocity and attenuation versus frequency in fine mortar containing 30% of 2.84 mm diameter expanded polystyrene balls (FM-P3).

FIG. 12. (Color online) Modeled (WT: continuous lines, and GSCM: mixed lines) and experimental (points) phase velocity and attenuation versus frequency in fine mortar containing 34.1% of 3 mm diameter glass beads and 17.1% of 2.84 mm diameter expanded polystyrene balls (FM-G3-P3).