An iterative effective medium approximation (IEMA) for wave dispersion and attenuation predictions in particulate composites, suspensions and emulsions

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In the present work we deal with the scattering dispersion and attenuation of elastic waves in different types of nonhomogeneous media. The iterative effective medium approximation based on a single scattering consideration, for the estimation of wave dispersion and attenuation, proposed in Tsinopoulos *et al.*, [Adv. Compos. Lett. **9**, 193–200 (2000)] is examined herein not only for solid components but for liquid suspensions as well. The iterations are conducted by means of the classical relation of Waterman and Truell, while the self-consistent condition proposed by Kim *et al.* [J. Acoust. Soc. Am. **97**, 1380–1388 (1995)] is used for the convergence of the iterative procedure. The single scattering problem is solved using the Ying and Truell formulation, which with a minor modification can accommodate the solution of scattering on inclusions in liquid. Theoretical results for several different systems of particulates and suspensions are presented being in excellent agreement with experimental data taken from the literature.
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I. INTRODUCTION

When a plane wave travels through a suspension of particles like particulate composites (solid particles in solids), liquid suspensions (solid particles in fluid), and emulsions (fluid inclusions in fluid), multiple scattering occurs and part of the incident energy is transferred to the scattered fields. Parameters such as the frequency of the incident wave, the relative position among the particles, the geometry of the particles and the material properties of both matrix and inclusions affect the amount of this energy. Thus, although matrix and particles can be nonattenuative, the amplitude of waves propagating through suspensions decays and the decay rate is frequency dependent. For a plane wave the decay of its amplitude is expressed via a frequency dependent exponential coefficient known as an attenuation coefficient. On the other hand, the size of the particles as well as the material mismatch between particles and surrounding medium imply that the dynamic behavior of the composite medium is strongly depended on the excitation frequency of the incident wave. Macroscopically this means that the phase velocity of a plane wave traveling through a suspension of particles is frequency dependent. This phenomenon is known in the literature as wave dispersion.

The quantitative determination of dispersion and attenuation of a plane wave, caused by a random distribution of inhomogeneities, is a problem which has been studied intensively either theoretically or experimentally by many investigators in the past. The first important theoretical work on the subject is that of Foldy[1] who, employing a configurational averaging procedure, derived a dispersion relation for scalar wave propagation through a medium containing isotropic scatterers. Later, Lax[2] extended the work of Foldy and proposed a new dispersion relation for multiple wave scattering by anisotropic scatterers. In both works the wave dispersion and attenuation was represented via a frequency dependent complex wave number expressed in terms of the particle concentration and the forward far field scattering amplitude taken from the solution of the single particle wave scattering problem. The results of Lax were further improved by Waterman and Truell,[3] Twersky,[4] Lloyd and Berry,[5] Varadan *et al.*[6] and Javanaux and Tomas[7] who derived dispersion relation expressed in terms of the particle concentration and the forward as well as the backward scattering amplitude of the single scattering problem inserting thus the contribution of the back-scattering to the multiple scattering process.

The above mentioned multiple scattering theories have been extensively exploited by many investigators in order to explain wave dispersion and attenuation observed in experiments dealing with wave propagation in nonhomogeneous fluids and solids. Here one can mention the representative works of Sayers and Smith,[8] Ledbetter and Datta,[9] Norris,[10] Anson and Chivers,[11] Shido *et al.*,[12] Lu and Liaw[13] and Challis *et al.*[14] for particulate composites, the works of Holmes *et al.*,[15] Mobley *et al.*,[16] Meulen *et al.*,[17] for elastic particles in liquid suspensions and the works of McClemens and Povey[18] and McClemens[19] for emulsions. In most of these articles, spherical inclusions are considered while the far field parameters of the single particle wave scattering problem, used in the dispersion and attenuation expressions, are mainly taken from the works of Epstein and Carhart[20] for emulsions, Allegra and Hawley[21] for elastic particles in a liquid continuum and Ying and Truell[22] for suspensions of

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solids in solids. Comparisons showed that, for cases of particulate composites with a significant mismatch between the physical properties of particles and matrix, the aforementioned multiple scattering theories predict well only for very low concentrations of particles (less than 10%), while their prediction efficiency, in terms of particles concentration, can be improved in cases of nonhomogeneous solids with small differences in the physical properties of the material constituents. On the other hand, the simple multiple scattering theories of Foldy,[1] Waterman and Truell[3] and Lloyd and Berry[5] enhanced by the Epstein and Carhart[20] and Allegra and Hawley[21] models, where besides the interaction of the spherical particle with the incident wave, heat transport phenomena between particles and surrounding medium are taken into account, provide reasonable predictions for liquid suspensions and emulsions with concentrations up to 20%. This is an expected result since, due to mode conversion, multiple scattering effects are more pronounced in solid than in liquid suspensions. Eventually, one can say here that none of the so far mentioned theories is able to provide acceptable wave dispersion and attenuation predictions for all the types of suspensions and for a wide range of particle concentrations and wavenumbers.

Besides the aforementioned fundamental multiple scattering procedures, many analytical semi-analytical and numerical models for predicting wave dispersion and attenuation in nonhomogeneous media have been proposed in the literature. Among them, the methodologies applied for both solid and liquid suspensions can be grouped into two categories. In the first category belong the works that provide dispersion and attenuation expressions by means of the Kramers–Kroning relations. Representative works are those of Beltzer *et al.*[23] and Beltzer[24] for particulate composites and the works of Temkin,[25] Ye[26] and Leander[27] for suspensions, while an excellent mathematical description and derivation of Kramers–Kroning relations can be found in the work of Weaver and Pao.[28] However, as it is mentioned in the book of Zhang and Gross[29] and noted in the paper of Temkin,[30] Kramers–Kroning relations provide satisfactory results only for low concentrations of particles while one of the quantities phase velocity and attenuation coefficient should be known independently. The second category concerns the self-consistent theories. According to these theories, the frequency dependent wave velocity and attenuation coefficient are evaluated through self-consistent expressions most of which are based on scattering parameters taken from the solution of the single scattering problem where the microstructure of the composite medium is immersed into an infinitely extended effective medium. The self-consistent or effective medium theories appear in the literature with different versions and procedures depending on the type of suspensions they applied. Thus, for particulate composites one can mention the self-consistent models of Talbot and Willis,[31] Sabina and Willis[32] and Devaney,[33] the effective medium approximations of Kerr[34] and Kanaun *et al.*,[35] the dynamic self-consistent effective medium approximations of Berryman,[36] Kim *et al.*[37] and Tsinopoulos *et al.*[38] and the incremental self-consistent approach of Anson and Chivers[11] and Biwa *et al.*[39] For liquid suspensions, one can mention the effective medium approaches of Anson and Chivers,[40] Hemar *et al.*,[41] Cowan *et al.*,[42] McClements *et al.*[43] and Hipp *et al.*[44,45] and the coupled-phase models of Harker *et al.*,[46] Atkinson and Kytomaa[47] and Evans and Attenborough.[48] Comparisons with experimental results have shown that the self-consistent models are those which are able to predict satisfactory the behavior of a wave pulse propagating within a dense distribution of particle-scatterers.

Recently, Kim *et al.*[37] presented a modified version of the coherent potential approximation,[49–52] in order to predict the speed and the coherent attenuation of an elastic wave propagating in a medium containing randomly distributed, solid spherical inclusions. The frequency dependent effective stiffness and density of the composite are obtained by solving a system of three nonlinear volume-integral equations in which, however, the interior dynamic displacement field of a single inclusion immersed in an infinitely extended effective medium must be known *a priori*. Although in their theory correlations among the scatterers are neglected, their results were in a good agreement with experimental observations.

Kanaun *et al.*[35] claim that the application of this effective medium scheme for wave propagation problems is questionable, since matrix and inclusions play quite different roles in the process of wave diffraction. However, scattering occurs due to the interaction of the incident wave with the randomly distributed particles. Thus, considering inclusions and matrix as scatterers the surfaces of which have opposite unit normal vector, the self-consistent hypothesis of Kim *et al.*[37] seems to be quite reasonable

Later, Tsinopoulos *et al.*[38] proposed an iterative effective medium approximation (IEMA) combining effectively the self-consistent model of Kim *et al.*[37] and the simple multiple scattering theory of Foldy.[1] In their work, the evaluation of the wave speed and attenuation coefficient was accomplished through a practical and simple iterative procedure avoiding thus the solution of complex nonlinear systems of equations such those required in the approximation of Kim *et al.* Moreover, comparing the estimations provided by the two methods, IEMA appears to be more efficient and accurate in cases of highly concentrated elastic mixtures.

Here, the IEMA of Tsinopoulos *et al.*[38] is properly modified and improved in order to predict well wave dispersion and attenuation in particulate composites, particle suspensions and emulsions. Our aim in the current work is twofold: first to develop a single theoretical model that predicts well wave dispersion and attenuation for all types of suspensions and second to provide an iterative computational scheme that for the case of spherical particles is simple and easily implemented. The present new version of IEMA combines the self-consistent model of Kim *et al.*[37] with the quasicrystalline approximation of Waterman and Truell.[3] Considering the effective material properties of the composite medium being the same with the static elastic ones proposed by Christensen,[53] properly modified for liquid mixtures, and satisfying the single scattering self-consistent condition of Tsinopoulos *et al.*,[38] the effective and frequency dependent dynamic density of the nonhomogeneous medium is evaluated. The complex value of the effective density in conjunction with the static effective stiffness of the composite medium determine both the velocity and the attenuation of an ultrasonic pulse propagating in the random particulate suspension. The single scattering problem is solved using the Ying and Truell[22] formulation, which with minor modification can accommodate solution of scattering problems dealing with inclusions suspended in liquid matrix. Several numerical results compared with experimental data taken from the literature demonstrate the IEMA efficiency on predicting wave dispersion and attenuation in all types of particle suspensions.

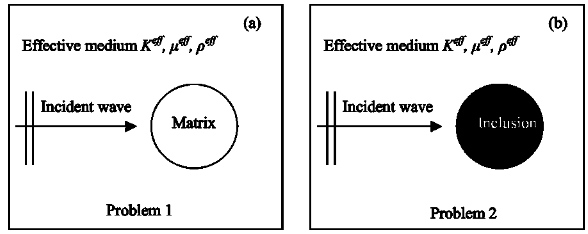

FIG. 1. A plane mean wave propagating in the effective medium and scattered by (a) a matrix inclusion (problem 1) and (b) a particle inclusion (problem 2).

II. THE IEMA FOR PARTICLE SUSPENSIONS

In this section, the IEMA proposed recently by Tsinopoulos *et al.*[38] and modified for the needs of the present work is presented.

The starting point of the IEMA is a self-consistent condition first considered in the coherent potential theory of Soven.[49] According to this theory, any wave propagating in a composite medium can be considered as a sum of a mean wave propagating in a medium having the dynamic effective properties of the composite and a number of fluctuating waves coming from the multiple scattering of the mean wave by the uniformly and randomly distributed material variations from these of the effective medium. On the average, the fluctuating field should be vanished at any direction within the effective medium, i.e.,

$$\langle \hat{\mathbf{k}} \cdot \tilde{\mathbf{T}} \cdot \hat{\mathbf{k}} \rangle = 0, \tag{1}$$

where $\langle \, \rangle$ denotes the average over the composition and the shape of the scatterers, $\tilde{\mathbf{T}}$ is a matrix corresponding to the total multiple scattering operator for the fluctuating waves and $\hat{\mathbf{k}}$ is the propagation direction of the mean wave. Equation (1) is well known as self-consistent condition and can be used to determine the dynamic effective properties of the composite material. However, due to the prohibitive computational cost of the evaluation of the operator $\tilde{\mathbf{T}}$ Soven[49] proposed, instead of Eq. (1), the use of the following simplified self-consistent condition:

$$\langle \hat{\mathbf{k}} \cdot \tilde{\mathbf{t}} \cdot \hat{\mathbf{k}} \rangle = 0, \tag{2}$$

with $\tilde{\mathbf{t}}$ being a single scattering operator coming from the diffraction of the mean wave by each composition, i.e. matrix and particles, embedded in an infinitely extended effective medium. Devaney[33] proved that Eq. (2) could also be written as a function of the far field scattering amplitudes in the forward direction. Thus, for identical homogeneous particles embedded in a homogeneous elastic or liquid matrix, Eq. (2) assumes the following form:

$$n_1 g^{(1)}(\hat{\mathbf{k}},\hat{\mathbf{k}}) + (1-n_1) g^{(2)}(\hat{\mathbf{k}},\hat{\mathbf{k}}) = 0, \tag{3}$$

where n_1 represents the volume fraction of the particles and $g^{(1)}(\hat{\mathbf{k}},\hat{\mathbf{k}})$, $g^{(2)}(\hat{\mathbf{k}},\hat{\mathbf{k}})$ are the forward scattering amplitudes taken by the solution of the two single wave scattering problems illustrated in Fig. 1. The solution of the single scattering problem is described in the next section.

According to the IEMA the self-consistent condition (3) is satisfied numerically through an iterative procedure, which can be summarized as follows.

Consider a harmonic elastic plane wave with circular frequency ω, either longitudinal (P) or transverse (SH or SV), traveling through the composite. Due to the presence of the particles, multiple scattering occurs and thereby the considered wave becomes both dispersive and attenuated and its complex wavenumber $k_d^{\text{eff}}(\omega)$ can be written as

$$k_d^{\text{eff}}(\omega) = \frac{\omega}{C_d^{\text{eff}}(\omega)} + i\,\alpha_d^{\text{eff}}(\omega), \tag{4}$$

with $C_d^{\text{eff}}(\omega)$ and $\alpha_d^{\text{eff}}(\omega)$ being the frequency dependent wave phase velocity and attenuation coefficient, respectively. The subscript d denotes either longitudinal ($d \equiv p$) or transverse ($d \equiv s$) wave.

Next, the composite material is replaced by an elastic homogeneous and isotropic medium with effective Lame' constants μ^{eff}, λ^{eff}, given by the static model of Christensen,[53]

$$\lambda^{\text{eff}} = \lambda_2 + \frac{n_1(\lambda_1-\lambda_2)\left(\lambda_2+\frac{4}{3}\mu_2\right)}{n_2(\lambda_1-\lambda_2)+\left(\lambda_2+\frac{4}{3}\mu_2\right)},$$
$$A\left(\frac{\mu^{\text{eff}}}{\mu_2}\right)^2 + 2B\left(\frac{\mu^{\text{eff}}}{\mu_2}\right) + C = 0. \tag{5}$$

Subscripts 1 and 2 indicate particle and matrix material properties, respectively, and A, B and C are functions of (μ_1, μ_2, n_1) given in the paper of Christensen.[53] Since the Lame' constant λ is usually referred to an elastic medium, in the present work where liquid suspensions and emulsions are considered the bulk modulus $K^{\text{eff}} = \lambda^{\text{eff}} + (2/3) gm^{\text{eff}}$ is used instead.

For the cases of a liquid matrix, the shear modulus, μ^{eff}, instead of being calculated through (5), is set to a very small value, since, even for high concentrations, the inclusions do not form an interconnected network that would effectively reinforce the shear rigidity of the mixture. In the present paper the shear modulus for all the considered liquid phases has been taken equal to 100 Pa.

In the first step of the IEMA, the effective density of the composite is assumed to be

$$(\rho^{\text{eff}})_{\text{step1}} = n_1 \rho_1 + (1-n_1)\rho_2. \tag{6}$$

Then, the effective wave number $(k_d^{\text{eff}})_{\text{step1}}$ is evaluated as straightforward through the relations

$$(k_p^{\text{eff}})_{\text{step1}} = \omega \left[\frac{3K^{\text{eff}} + 4\mu^{\text{eff}}}{3(\rho^{\text{eff}})_{\text{step1}}} \right]^{-1/2}, \quad (7)$$

for a P-wave and

$$(k_s^{\text{eff}})_{\text{step1}} = \omega \left[\frac{\mu^{\text{eff}}}{(\rho^{\text{eff}})_{\text{step1}}} \right]^{-1/2}, \quad (8)$$

for a shear wave, respectively.

In the sequel, utilizing the material properties obtained from the first step, the two single wave scattering problems illustrated in Fig. 1 are solved. The solution of these problems is accomplished analytically from the matrix notation of the Ying and Truell formulation, as will be explained in Sec. III. Combining the evaluated forward scattering amplitudes $g_d^{(1,2)}(\hat{\mathbf{k}},\hat{\mathbf{k}})$, according to the self-consistent condition (3), i.e.,

$$g_d(\hat{\mathbf{k}},\hat{\mathbf{k}}) = n_1 g_d^{(1)}(\hat{\mathbf{k}},\hat{\mathbf{k}}) + (1-n_1) g_d^{(2)}(\hat{\mathbf{k}},\hat{\mathbf{k}}), \quad (9)$$

and making use of the dispersion relation proposed by Waterman and Truell,[3] one obtains the new effective wave number of the mean wave,

$$[(k_d^{\text{eff}})_{\text{step2}}]^2 = [(k_d^{\text{eff}})_{\text{step1}}]^2 + \frac{3n_1 g_d(\hat{\mathbf{k}},\hat{\mathbf{k}})}{a^3} + \frac{9n_1^2(g_d^2(\hat{\mathbf{k}},\hat{\mathbf{k}}) - g_d^2(\hat{\mathbf{k}},-\hat{\mathbf{k}}))}{4k^2a^6}, \quad (10)$$

where a is the radius of the smallest sphere including the particle.

The new complex wave number $(k_d^{\text{eff}})_{\text{step2}}$ of the mean wave propagating through the composite medium is the departure point of the second step. Keeping the same static material properties (5) for the effective medium and utilizing relations (7) and (8) for longitudinal and transverse incidence, respectively, one calculates the new effective density of the host medium $(\rho^{\text{eff}})_{\text{step2}}$, which due to $(k_d^{\text{eff}})_{\text{step2}}$ is now complex. Considering the new material properties $\lambda_{\text{eff}}, \mu_{\text{eff}}$ and $(\rho^{\text{eff}})_{\text{step2}}$, the two single wave scattering problems depicted in Fig.1 are solved again and the procedure is repeated until the self-consistent condition (3) is satisfied. This means that $(k_d^{\text{eff}})_{\text{step}(n-1)} = (k_d^{\text{eff}})_{\text{step}(n)}$. Finally, the evaluated k_d^{eff} in conjunction with Eq. 4 determines the frequency dependent, effective velocity $C_d^{\text{eff}}(\omega)$ and the attenuation coefficient $a_d^{\text{eff}}(\omega)$ of the propagating wave. The whole procedure is summarized in the flow chart of Fig. 2.

In the just described procedure, a point that needs further discussion is the use of the complex density throughout the iterations of the IEMA. From a physical point of view, one can say that the choice of using the density as the main parameter controlling the material properties of a particle suspension seems to be realistic, since both dispersion and attenuation are dynamic properties of the considered composite medium. On the other hand the idea of a complex density is not something new in the literature. As a representative example one can mention the works of Petculescu and Wilen,[54] Lee *et al.*[55] and Pan and Horne[56] dealing with sound

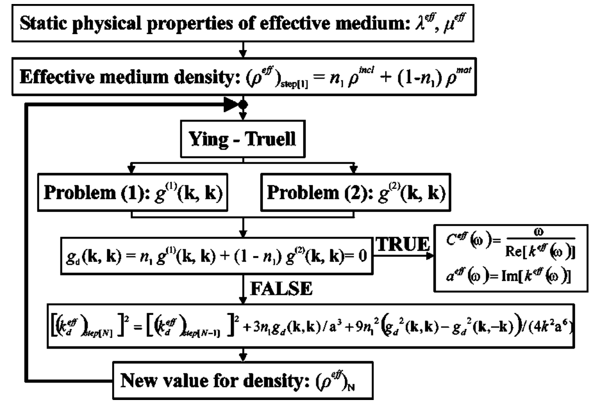

FIG. 2. A schematic representation of IEMA.

propagation and fluid flow in porous media and that employ complex densities in their frequency domain analysis. Usually, the imaginary and real part of a complex density come from the frequency domain transformation of first and second order time derivatives, respectively, involved in the differential operator of the problem. Thus, beyond the different type of explanations being available in the literature, the real and imaginary part of a complex density are directly related to the kinetic and the absorbing energy of the medium, respectively. This could explain why in the proposed here iterative methodology the complex density is responsible for the final evaluation of the frequency dependent velocity and attenuation coefficient of the particulate composite medium.

It should be also mentioned that an alternative IEMA procedure would be the consideration of a constant density [Eq. (6)] for all the steps and the use of either the complex values of the bulk modulus evaluated from Eq. (7) when longitudinal waves propagate through the composite medium, or the complex shear modulus obtained from Eq. (8) when shear waves are considered. However, although the two procedures seem to be equivalent, the use of the complex modulus instead of the complex density leads to dispersion and attenuation predictions that in many cases are in poor agreement with experimental observations. On the contrary and as it is evident in the sections after next, the use of the complex density in the IEMA procedure provides results being in a very good and sometimes in excellent agreement with the available experimental data.

III. FORMULATION AND SOLUTION OF THE SINGLE SCATTERING PROBLEM

In this section the formulation and solution of the single scattering problem is briefly described. The present approach is based on the Ying and Truell formulation[22] considering scattering of a plane wave on an elastic sphere embedded in an infinite elastic matrix. The subject of scattering on a spherical obstacle has been discussed extensively in literature so only general guidelines will be presented here.

When a compressional wave impinges on the particle it gives rise to both compressional and shear waves inside the particle, as well as the scattered compressional and shear waves outside the particle. Expressions for each of these waves are equated using the boundary conditions at the surface of the particle, yielding four equations with four unknown scattering coefficients, A_n, B_n, C_n and D_n. In the present formulation temperature and heat transfer effects are not included. The equations, concerning namely the continuity of the normal and tangential velocity component as well as the continuity of the normal and tangential stress component are as follows:

$$A_n k_1 \mathrm{a} h_{n+1}(k_1\mathrm{a}) + B_n n \kappa_1 \mathrm{a} h_{n+1}(\kappa_1\mathrm{a}) - C k_2 \mathrm{a} j_{m+1}(k_2\mathrm{a}) - D_n n \kappa_2 \mathrm{a} j_{m+1}(k_2\mathrm{a}) = (-i)^{n-1}(2n+1)\frac{1}{k_1}[k_1 \mathrm{a} j_{m+1}(k_1\mathrm{a})], \tag{11}$$

$$A_n h_n(k_1\mathrm{a}) - B_n[(n+1)h_n(\kappa_1\mathrm{a}) - \kappa_1\mathrm{a}h_{n+1}(\kappa_1\mathrm{a})] - C_n j_m(k_2\mathrm{a}) + D_n[(n+1)j_n(\kappa_2\mathrm{a}) - \kappa_2\mathrm{a}j_{n+1}(\kappa_2\mathrm{a})] = (-i)^{n-1}(2n+1)\frac{1}{k_1}j_m(k_1\mathrm{a}), \tag{12}$$

$$A_n[(\kappa_1\mathrm{a})^2 h_n(k_1\mathrm{a}) - 2(n+2)k_1\mathrm{a}h_{n+1}(k_1\mathrm{a})] + B_n n[(\kappa_1\mathrm{a})^2 h_n(\kappa_1\mathrm{a}) - 2(n+2)\kappa_1\mathrm{a}h_{n+1}(\kappa_1\mathrm{a})] - C_n p[(\kappa_2\mathrm{a})^2 j_n(k_2\mathrm{a}) - 2(n+2)k_2\mathrm{a}j_{n+1}(k_2\mathrm{a})] - D_n p n[(\kappa_2\mathrm{a})^2 j_n(\kappa_2\mathrm{a}) - 2(n+2)\kappa_2\mathrm{a}j_{n+1}(\kappa_2\mathrm{a})] = (-i)^{n-1}(2n+1)\frac{1}{k_1}[(\kappa_1\mathrm{a})^2 j_m(k_1\mathrm{a}) - 2(n+2) \times k_1\mathrm{a}j_{m+1}(k_1\mathrm{a})], \tag{13}$$

$$A_n[(n-1)h_n(k_1\mathrm{a}) - k_1\mathrm{a}h_{n+1}(k_1\mathrm{a})] - B_n\left[\left(n^2 - 1 - \frac{\kappa_1^2\mathrm{a}^2}{2}\right)h_n(\kappa_1\mathrm{a}) - \kappa_1\mathrm{a}h_{n+1}(\kappa_1\mathrm{a})\right] - C_n p[(n-1)j_n(k_2\mathrm{a}) - k_2\mathrm{a}j_{n+1}(k_2\mathrm{a})] + D_n p\left[\left(n^2 - 1 - \frac{\kappa_2^2\mathrm{a}^2}{2}\right)j_n(\kappa_2\mathrm{a}) - \kappa_2\mathrm{a}j_{n+1}(\kappa_2\mathrm{a})\right] = (-i)^{n-1}(2n+1)\frac{1}{k_1}[(n-1)j_m(k_1\mathrm{a}) - k_1\mathrm{a}j_{m+1}(k_1\mathrm{a})], \tag{14}$$

where k_1 and k_2 are the longitudinal wavenumbers in the matrix and inclusion, respectively, κ_1 and κ_2 are the shear wavenumbers in the matrix and inclusion, respectively, $p = \mu_2/\mu_1$, a is the particle radius, and j_n and h_n are the spherical Bessel and Hankel functions.

In case the modeling concerns a problem of scattering on particles suspended in liquid, the equations can be derived by a limiting process $(\mu_2 \to 0)$.[59] As it is already mentioned in the present work the shear modulus for any liquid phase was taken to be 100 Pa.

In order to calculate velocity and attenuation for a given frequency the equations must be solved for the scattering coefficients for each value of n. In the case of the scattered longitudinal wave, the A_n coefficients are of interest. This system of equations, in matrix notation, is solved by a Matlab routine using standard inversion command. Therefore, the forward scattering amplitude, g, can be calculated through

$$g(0) = \frac{1}{ik_1}\sum_{n=0}^{\infty}(2n+1)A_n, \quad g(\pi) = \frac{1}{ik_1}\sum_{n=0}^{\infty}(-1)^n(2n+1)A_n. \tag{15}$$

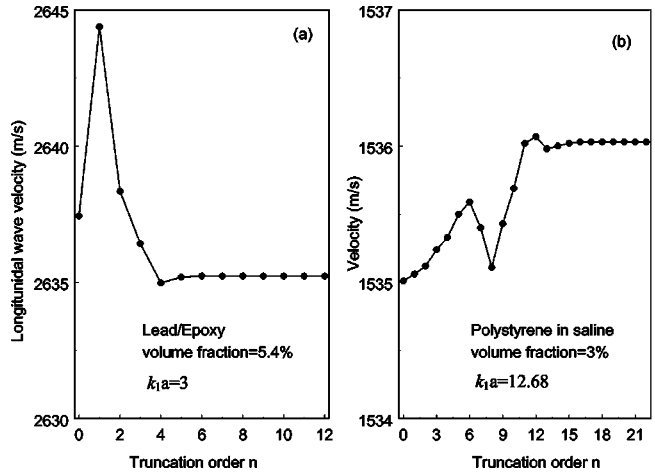

FIG. 3. (a) Velocity vs order n for 5.2% by volume composite of 660 μm radius lead spheres in epoxy. (b) Velocity vs order n for 3% by volume suspension of 152 μm radius polystyrene spheres in water.

The appropriate order of n has been shown by O'Neil *et al.*[60] to be roughly equal to the dimensionless wavenumber, $k_1 a$, meaning that for higher frequency bands as the particle radius rises significantly compared to the wavelength, and in order to have a reliable calculation of the scattering amplitude and therefore velocity and attenuation, more scattering terms must be summed in Eq. (15).

Indeed, this is evident throughout the present work as can be seen in Fig. 3. There, two indicative examples concerning the order of n, necessary for the convergence of velocity via the Waterman and Truell dispersion relation are depicted. In Fig. 3(a) where a case of a lead/epoxy particulate composite is considered, it is seen that for frequency 1.9 MHz corresponding to $k_1 a = 3$, for particle radius 660 μm, the velocity obtains a constant value after about $n = 6$. In Fig. 3(b) the medium is a polystyrene in the saline suspension and the appropriate number of n for convergence is 16 while the $k_1 a$ equals 12.5 (radius 152 μm and frequency 20 MHz). It is seen that generally velocity and attenuation converges for order $k_1 a + 3$ while thereafter no detectable change is mentioned. Therefore n was set equal to the integer of $k_1 a + 7$ for the needs of the present study.

IV. RESULTS AND DISCUSSION

In this section the prediction capability of the IEMA is examined. Although it has been proven to yield accurate results for particulate composites[38] for volume concentrations as high as 50%, herein the efficiency of IEMA is addressed also for liquid matrix systems. Calculations are carried out for several cases of systems for which experimental data are

TABLE I. Material properties.

Material	Cp (m/s)	Cs (m/s)	λ (Gpa)	μ (Gpa)	ρ (Kg/m^3)	Fig.
Iron	5941	3251	110.99	82.86	7840	4(a)
PMMA	2669	1305	4.37	2.00	1175	
TiC	10000	6200	113.28	188.36	4900	4(b)
Epoxy	2523	976	5.58	1.19	1250	
Lead	2210	860	38.48	8.36	11300	5
Epoxy (828z)	2640	1200	4.92	1.73	1202	
Al7091	6305	3066	59.5	26.7	2840	6
SiCp	12210	7707	100.0	196.0	3300	
Glass	5280	3240	17.15	26.16	2492	7
Epoxy (3012)	2541	1161	4.44	1.59	1180	
Polystyrene	2337	1098	3.21	1.27	1053	8, 9(b), 10
Water	1500	—	2.250	–	1000	8, 9(b), 10, 12, 13, 14(b)
Glass	6790	4167	27.36	41.76	2405	9(a)
ATB	1026	—	2.49	–	2365	
Glass	5600	3400	20.6	28.9	2500	11
Glycerol–water mixture	1840	—	4.063		1200	11
Xylene	1320	—	1.513	–	868.2	12
Bromoform	900	—	2.341	–	2890	13(a)
Benzene	1320	—	1.5263	–	876	13(b), 14(a)
Water/glycerine	1711	—	3.232	–	1104	14(a)
Carbon tetrachloride	968	—	1.536	–	1640	14(b)

available in the literature. The material properties of the constituent phases are summarized in Table I along with the corresponding figure number where experimental and theoretical curves are depicted. Although water and polystyrene takes part in more than one measurement, the properties do not exhibit remarkable differences from one case to other; therefore they are mentioned once.

A. Particulate composites

The first material studied is an Iron/PMMA composite. In Fig. 4(a) comparison between measured and calculated longitudinal velocity is depicted for a monochromatic wave ($k_1 a = 0.06$). The experimental data are obtained by the work of Piche and Hamel.[57] As observed the agreement is excellent while the Waterman–Truell model, as expected, predicts well only for low concentrations.

The other case, Fig. 4(b), concerns a titanium carbonate (TiC) in epoxy composite at the dimensionless frequency $k_1 a = 0.02$. It is obvious that the agreement for almost all volume fractions between experimental velocity[11] and IEMA predictions is very good. In this figure, the predictions made by the Waterman and Truell approach, without applying the iterative procedure, are also supplied. It is seen that the agreement is good but for high volume fractions the use of the self-consistent relation seems to more closely follow the trend at higher volume fractions.

The next case under consideration is a lead/epoxy (Epon 828z) composite with spherical particles of radius 660 μm and volume fractions 26% and 52%; see Figs. 5(a) and 5(b), respectively. The theoretical results are compared again to those taken directly from the Waterman–Truell dispersion relation. Although the discrepancy between IEMA results and experiment (taken from the work of Kinra and Rousseau[58]) seems to increase with volume content, it can be said that, qualitatively, the results are in good agreement. An

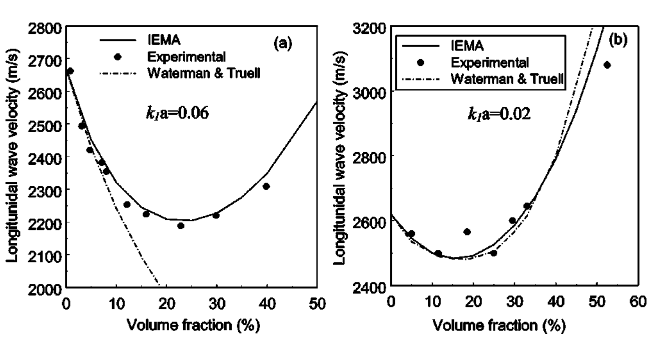

FIG. 4. A comparison between predicted and measured longitudinal velocity for (a) an iron/PMMA composite (experimental data from Ref. 52) and (b) for a titanium carbonate in epoxy composite (experimental data from Ref. 11).

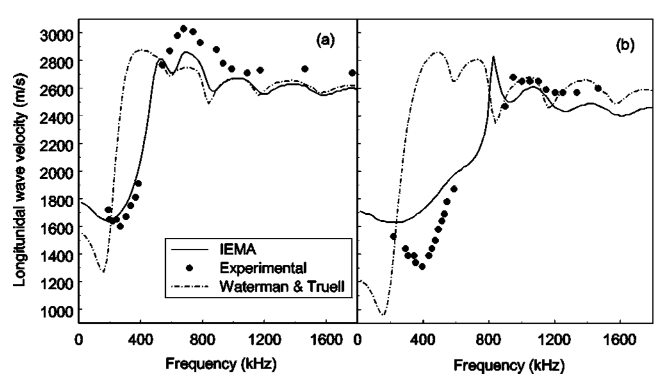

FIG. 5. A comparison between predicted and measured (Ref. 58) longitudinal velocity for a 660 μm radius lead spheres in an epoxy composite with volume fraction (a) 26% and (b) 52%.

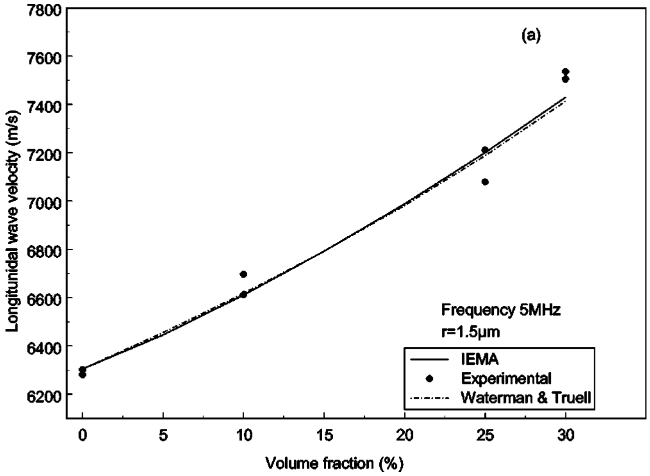

FIG. 6. A comparison between predicted and measured (Ref. 13) longitudinal velocity for a SiCp in aluminum composite, matrix type 7091.

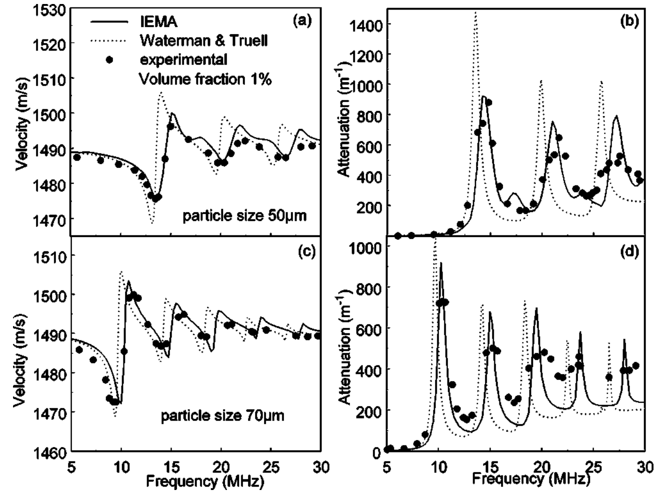

FIG. 8. A comparison between the predicted and measured (Ref. 16) sound velocity of a suspension of 1% polystyrene spheres in water with radius (a) 50 μm and (c) 70 μm and corresponding attenuation (b) and (d).

important conclusion drawn by this figure is that the IEMA predicts the shift of the lowest and highest resonance frequencies to higher values as the volume fraction increases. The position of the resonant frequencies can be found in the diagrams of Fig. 5 since at these frequencies the velocity obtains maximum values.

Figure 6 depicts the longitudinal wave velocity of aluminum (Al) matrix composites containing silicon carbide (SiCp) particles. The increase of velocity with volume fraction is apparent for both cases and predicted values are quite close to the experimental ones concerning the random orientation of the particles. After microstructural characterization of the material,[13] the average SiCp size varied approximately from 2 to 4 μm. For the theoretical predictions the diameter was considered to be 3 μm.

The last particulate composite case concerns the attenuation of the glass/epoxy (Tra-cast 3012) system. The attenuation measurements carried out by Kinra *et al.*[61] for a 45% volume content of glass and the corresponding predictions of IEMA are presented in Fig. 7. It is apparent that the IEMA follows closely the experimental data for the frequency range tested, while the Waterman and Truell approach seems suitable only for low frequencies.

B. Elastic–liquid suspensions

Liquid suspension modeling does not require a much different approach. As mentioned above, since liquids do not support shear waves, the shear modulus of the matrix obtained values very small (100 Pa).

The first case studied is a 1% by volume polystyrene in water suspension with particle size 50 μm interrogated experimentally[16] in the frequency band 3–30 MHz by means of ultrasonic spectroscopy. Overall, the predicted shape of the phase velocity and attenuation curves tracks the experimental results closely as seen in Figs. 8(a) and 8(b), respectively. The peaks and nadirs of the IEMA model coincide with the experimentally measured ones for both velocity and attenuation being closer than the original Waterman and Truell dispersion relation.

Another case of interest lies in Fig. 9(a). There, the velocity of glass in acetylene tetrabromide-benzene (ATB) is

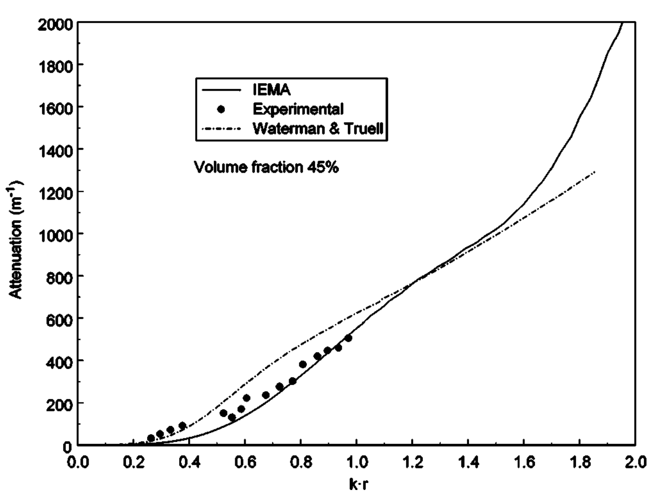

FIG. 7. A comparison between the predicted and measured (Ref. 61) longitudinal attenuation for a glass/epoxy (Tra-cast 3012) composite.

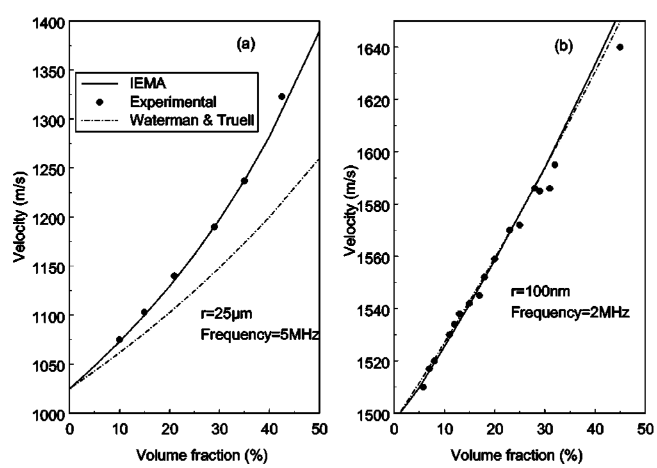

FIG. 9. A comparison between the predicted and measured sound velocity of a suspension of (a) glass in ATB (experimental data from Ref. 18) and (b) polystyrene in water (experimental data from Ref. 15).

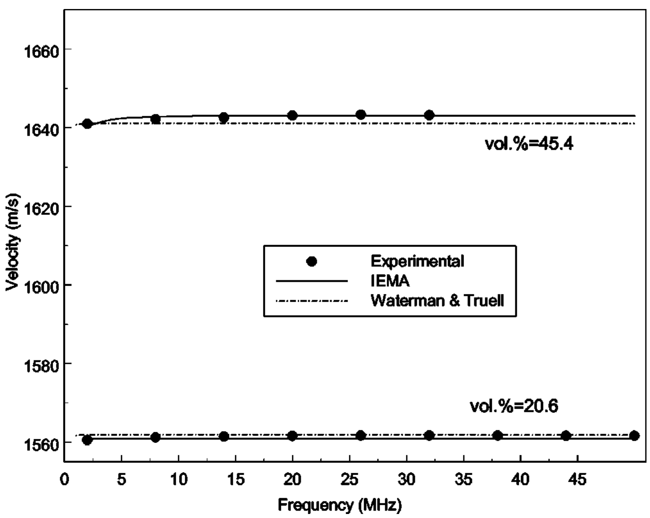

FIG. 10. A comparison between the predicted and measured (Ref. 15) sound velocity of a 308 nm polystyrene spheres in water suspension for different volume contents.

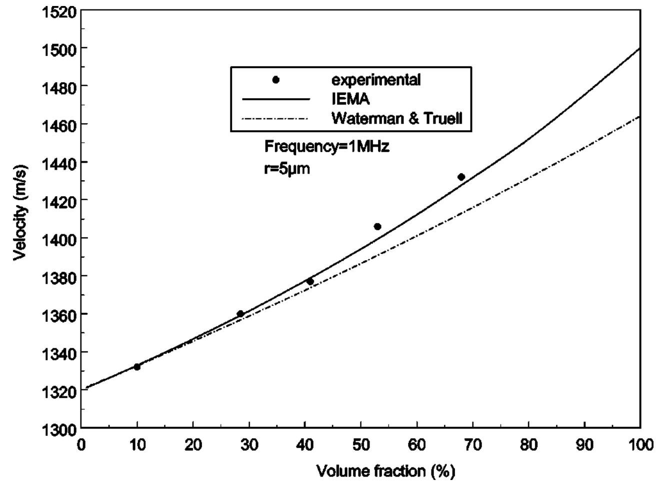

FIG. 12. A comparison between the predicted and measured (Ref. 18) sound velocity of water in a xylene emulsion.

depicted vs the volume content. The frequency applied experimentally is 5 MHz while the particle radius is 12.5 μm, resulting in $k_1 a = 0.3829$. The experimental data is due to McClements and Povey[18] and they are in excellent agreement with IEMA predictions.

In Fig. 9(b) another case of polystyrene in water is examined concerning the effect of the volume fraction in velocity at 2 MHz. In such systems, due to the low-density contrast between the two phases thermal effects are expected to be dominant.[15] However, although the present formulation omits such effects, theoretical predictions are very close to experimental data.[15]

The same work of Holmes *et al.*[15] contains interesting comparisons between the dispersive behavior of different particle volume fraction suspensions sharing though the same particle size. In Fig. 10 the experimentally observed dispersion of 20.6% and 45.5% with a particle size of 308 nm between 2 and 50 MHz is depicted. It is apparent that both the IEMA and the Waterman–Truell model predict well for the present case.

The last case of elastic in a liquid suspension presented herein concerns a suspension of monodisperse (radius 0.438 mm) glass beads in a 75% glycerin–25% water mixture. The properties of the suspended and continuum media exhibit large mismatch (the sound velocity of fluid is 1840 m/s while of glass beads 5600 m/s) resulting in strongly scattering behavior. IEMA succeeds in predicting very well the experimental behavior[42] up to about 5 MHz tested for the cases of 34% and 45% volume content of glass as seen in Figs. 11(a) and 11(b), respectively.

C. Liquid–liquid emulsions

Apart from the suspension of elastic particles in fluid, a separate category can be assumed for the liquid–liquid emulsions. Ultrasonic parameters of such systems as velocity and attenuation can also here be very closely predicted using $\mu = 100$ Pa for both liquids and following the same iterative procedure. All experimental data concerning this section are taken again from McClements and Povey.[18]

In Fig. 12 a water in xylene emulsion is described with droplet size 5 μm, measured at the frequency of 5 MHz for a wide range of volume fractions. In the next Fig. 13 two cases of water based emulsions are presented. In Fig. 13(a) the dispersed phase is bromoform and in Fig. 13(b) it is benzene. The droplet size is 3 μm for the first case and 8 μm for the second while the frequencies used are 3 and 2 MHz, respectively. As can be seen, the increase of the dispersed liquid

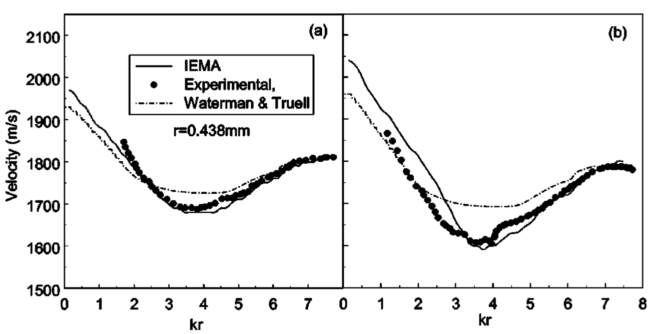

FIG. 11. A comparison between the predicted and measured (Ref. 42) sound velocity of a glass beads in glycerol–water mixture suspension for volume fraction (a) 34% and (b) 45%.

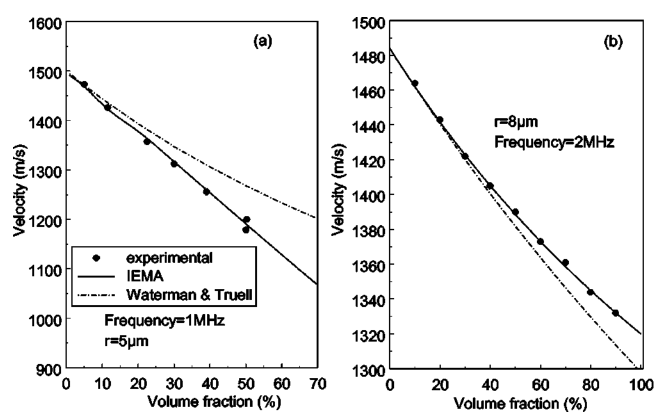

FIG. 13. A comparison between the predicted and measured (Ref. 18) sound velocity of (a) bromoform in water and (b) benzene in water emulsion.

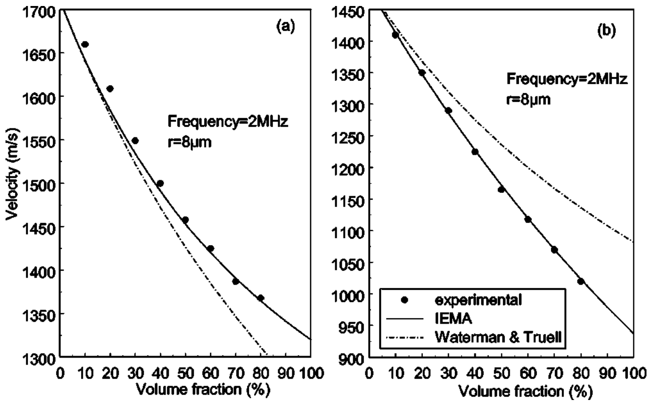

FIG. 14. A comparison between the predicted and measured (Ref. 18) sound velocity of benzene in water/40% glycerine and (b) carbon tetrachloride in water emulsion.

content causes a certain decrease in sound velocity, which is predicted exactly by the IEMA present herein.

The cases of the last figure concern a benzene in water/40% glycerin emulsion, see Fig. 14(a), and a carbon tetrachloride in water emulsion, Fig. 14(b). It is seen that the decrease in velocity with the increase of a droplet content is very closely predicted using the iterative procedure on the Ying and Truell formulation described herein. The properties of all different phases of the emulsions can be found in Table I.

V. CONCLUSIONS

A recently introduced iterative methodology for the quantitative estimation of wave dispersion and attenuation due to scattering is described here. Although successfully tested for particulate composites,[38] in this work its effectiveness is examined additionally on suspensions of solids in liquid and liquid in liquid emulsions systems. The obtained theoretical curves concerning velocity and attenuation predict very closely the experimental results, regardless of the nature of the phases. The elastic properties of the effective medium are given by the static mixture model of Christensen[53] while for the case of liquid matrix the shear rigidity is taken as 100 Pa. The dynamic behavior of the system is characterized by the complex effective material density, calculated through the iterative procedure discussed above. The IEMA being simpler than the one proposed by Kim *et al.*[37] is very powerful even for high volume fractions where it provides predictions close to experimental observations. The IEMA can be used as a practical tool for wave dispersion and attenuation prediction being very useful for applications in a nondestructive evaluation. Taking into consideration thermal effects is expected to improve the accuracy of dispersion and attenuation predictions and is proposed as a task for future work.

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