MODELING OF ULTRASONIC ATTENUATION IN UNI-DIRECTIONAL FIBER REINFORCED COMPOSITES COMBINING MULTIPLE-SCATTERING AND VISCOELASTIC LOSSES

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ABSTRACT. A model coupling viscoelastic and multiple-scattering losses is developed to predict ultrasonic attenuation in unidirectional fiber reinforced composite of high fiber volume fraction. Complex-valued stiffness constants accounting for viscoelasticity are inserted in classical multiple-scattering theory. Waves of various polarities (SH, SV, L) relatively to the fiber direction are considered. SH waves only require a scalar treatment, whereas the others require a vector treatment accounting for mode-conversions. Comparisons of predicted attenuation coefficients with experimentally measured ones validate the model.

INTRODUCTION

Unidirectional fiber reinforced layers enter in many manufacturing processes of composite materials. The geometry of such a layer leads to anisotropic symmetry of elastic characteristics (transversely isotropic). This property may be most useful for certain parts. By combining unidirectional layers of various orientations, materials of more complicated symmetries can be produced.

There is an increasing need for the development of ultrasonic nondestructive testing (UT) methods for inspecting parts made of composite materials following the increasing use of such materials in the industry (*e.g.*, aircraft and aerospace industries). In order to design new UT methods or to demonstrate performances of existing ones, simulation tools are very helpful [1]. Since attenuation plays an important role in actual testing of composites, models on which these tools are based must take into account the various attenuation phenomena encountered.

Among the unidirectional fiber reinforced composite materials, those made of carbon fibers in a resin matrix constitute a very common class. As far as UT is concerned, this class of material is quite challenging. At first, their anisotropic nature leads to complex wave behavior, as it is the case for unidirectional fiber reinforced composites in general. Moreover, for this class of composite materials, ultrasonic waves are highly attenuated: *i)* the resin matrix exhibits viscoelastic properties, *ii)* part of the energy is lost, scattered by fibers. Eventually, it seems obvious that both phenomena are intrinsically coupled.

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In the literature, one can find many works on modeling viscoelastic losses (see for example Deschamps and Hosten [2]) as well as losses due to fiber scattering (see for example Varadan *et al* [3]). As far as fiber scattering is concerned, some works deal with theories for single fiber scattering, some others take into account multiple-scattering phenomena. Biwa [4] developed a model combining losses due to both independent scattering by fibers (single scattering theory) with viscoelastic effects where the two phenomena simply superimpose. To the authors' knowledge, no work has been published where mutiple scattering and viscoelastic losses are simultaneously accounted for.

Our attempt here is to develop such a theoretical point-of-view since intuitively, the two phenomena shall be coupled to a certain extent. Experimental evidence of its validity will be shown by comparing predictions with existing measurements [5].

The present paper is therefore organized as follows. In the first part, a theoretical model coupling viscoelastic losses with losses due to multiple-scattering by fibers is derived. The second part is dedicated to the discussion of theoretical results. In particular, we compare theoretical results obtained with this new model to existing theoretical results recently predicted in [4]. The third part of the paper is dedicated to an experimental validation of the present model for various wave polarizations relatively to the fiber direction and materials with various fiber volume fractions.

THEORY

One of our aims in deriving the present model was to describe a two-phase unidirectional fiber-reinforced layer as an anisotropic and attenuating material homogeneous at the wavelength scale for further simulations [6]. Considering the typical diameter of fibers (7 µm), the typical fiber volume fraction (65%) and the typical frequency range of ultrasonic testing of one composite of interest [6], the wavelength scale is such that viscosity, multiple-scattering and their possible coupling appear as a (homogeneous) global phenomenon. Under such an assumption, the effect on wave amplitude of the overall attenuation phenomena maybe written as a simple frequency-dependent filter as

$$\exp\left[-\alpha_0 \omega^p d\right], \tag{1}$$

such a writing allowing quite simple computer implementation in a more general propagation model. In this expression, α_0 and p are real and positive valued, ω denotes the angular frequency and d is the distance of propagation in the medium considered.

Correlatively, one other aim was to predict the phase velocities for the various possible wave polarizations as functions of the frequency and to study their possible dispersion. The model considers ultrasonic waves of various polarities defined relatively to the fiber direction. To simplify, only the cases of waves propagating perpendicularly to the fibers will be considered. In what follows (see Fig. 1), SH (respectively SV) denotes shear waves with a polarization vector parallel (respectively perpendicular) to the fiber direction

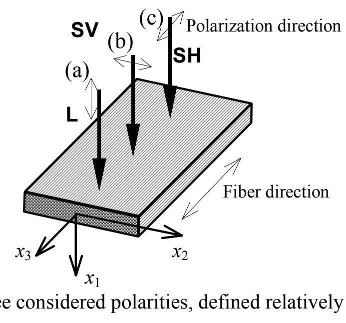

FIGURE 1. Description of the three considered polarities, defined relatively to the fiber direction.

and L denotes compression waves polarized along the wave vector and propagating perpendicularly to the fiber direction. The matrix is considered as an isotropic medium and the fiber, as a transversely isotropic medium whose axis of symmetry is parallel to the fiber orientation.

Therefore, for symmetry reasons, the SH case only requires a scalar treatment since no mode-conversion can occur, whereas the two others require a full vector treatment accounting for mode-conversion phenomena in the fiber scattering process.

Viscoelastic Losses

In the literature, the rheological approach is often used to model viscoelastic losses. Such an approach for a polymer matrix results in an attenuation filtering effect where the angular frequency [see Eq. (1)] is squared, under the classical low viscosity approximation [8]. However, there are many experimental evidences for the case of viscous material (*e.g.* epoxy resin) showing a linear behavior [9], rather. Correlatively, measurements show that over a frequency range typical of that used in UT, phase velocity undergoes negligible dispersion (*i.e.*, is almost independent of the frequency).

These experimental observations allow us (following Biwa *et al* [7]) to describe the viscoelastic matrix as an isotropic medium with complex-valued Lamé's coefficients, of which both the real and the imaginary parts are frequency-independent.

In the case of the epoxy resin considered in the following theoretical predictions compared to experimental measurements, the coefficients taken into account are given by $\lambda = 4.45 - i\,(0.027)$ and $\mu = 1.58 - i\,(0.128)$ (GPa), as measured ultrasonically by Biwa [5].

Multiple-Scattering Theories

The problem of multiple scattering of waves by a set of scatterers is a classical problem of theoretical physics. In the case of elastic wave scattering, solutions to this problem were derived in the early 1950's. Some of the theories were developed in order to describe a two-phase material composed of fibers in a matrix as a homogeneous material. Among these theories, the quite recent one developed by Yang and Mal [10] known as the generalized self-consistent method relies on the description of the heterogeneous medium as being constituted of three cylindrical phases (scattering by fibers being considered). This three-phase description is combined with the classical multiple-scattering theory originally derived by Waterman and Truell [11] for spherical scatterers modified to account for the cylindrical symmetry. Similar modifications could be made to enable the approach to deal with other geometry of scatterer.

Fundamentally, the idea of a three-phase modeling approach is to describe the homogeneous effective medium as three concentric cylinders as shown on Fig. 1. The center medium is nothing but the fiber itself. The intermediate one shares the same elastic properties as those of the matrix. The surrounding medium is an artificial synthetic medium

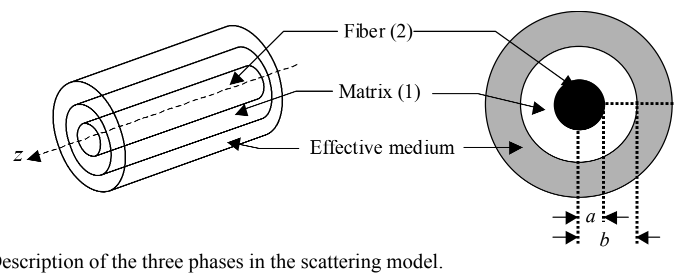

FIGURE 2. Description of the three phases in the scattering model.

that mathematically would represent the overall effective homogeneous medium to be determined. As far as boundary conditions are concerned, equations of continuity for stresses and displacements across the fiber-matrix interface write the same way as if there were only two phases. Radius a is that of the fiber. Radius b is a parameter that depends on the fiber volume fraction ϕ and is related to the average free path of scattered waves in the matrix. It depends on a and ϕ and equals $b = a/\sqrt{\phi}$. The density of the effective medium is a simple function of ϕ and of densities ρ_m and ρ_f of the matrix and the fiber. It is given by

$$\langle \rho \rangle = (1-\phi)\rho_m + \phi \rho_f . \tag{2}$$

The complex-valued wavenumber denoted by k is the solution of the problem in hands and will describe the wave behavior in the effective medium. Its real part is related to the frequency-dependent phase velocity and its imaginary part is the frequency-dependent attenuation coefficient. Wavenumber k writes

$$\langle k \rangle = \frac{\omega}{V(\omega)} + i\alpha(\omega) . \tag{3}$$

In Eqs. (2-3), the notation $\langle \circ \rangle$ means that the quantity is that of the effective medium. In what follows, it will be omitted and $\langle k \rangle$ will be simply denoted by k.

The scope of the present paper is not to recall the whole derivation of the model proposed by Yang and Mal [10]. Readers interested in this detailed description are referred to Ref. 10. Our aim here is to recall the main stages of this theory that will be used further and add a more detailed discussion about its numerical implementation.

The case of SH waves that only requires a scalar treatment and that of L and SV waves that requires a vector one to deal with mode-conversion phenomena at interfaces are considered separately. In both cases, the derivation leads to the resolution of a system made up of equations of continuity for the stress and the particle displacement at the two interfaces ($r=a$ and $r=b$ as shown on Fig. 2). One (SH case) or two (L and SV case) supplementary equations are required to solve the problem of finding the complex-valued wavenumbers associated to these waves. These supplementary equations relate to the multiple-scattering phenomenon. They were derived by Yang and Mal, as mentioned above, from Waterman and Truell [11] theory of multiple-scattering.

Let us consider here the simple SH case to describe how the supplementary equation given by Eq. (4) is solved in practice. Waterman and Truell give the relation

$$\left(\frac{k}{k_1^2}\right)^2 = \left[1 - \frac{2in_0 f_s(0)}{k_1^2}\right]^2 - \left[\frac{2in_0 f_s(\pi)}{k_1^2}\right]^2 , \tag{4}$$

where k_1 is the SH wavenumber in the matrix, $f_s(\theta)$ is related to the far-field amplitude of a wave scattered by a fiber and propagating in the θ-direction, n_0 is the number of scatterers per unit area. Applied to the three-phase model, the far-field expression is now related to the effective medium so that k_1 is substituted for k in Eq. (4). This leads to an implicit expression since the function $f_s(\theta)$ depends on the unknown k. One gets

$$1 = \left[1 - \frac{2in_0 f_s(0)}{k^2}\right]^2 - \left[\frac{2in_0 f_s(\pi)}{k^2}\right]^2 . \tag{5}$$

Due to its implicit nature, this equation is then solved by means of an iterative scheme. Numerically, the scheme is initiated by taking for k initial value that corresponds to a propagation into the matrix material, k_1. The global system made up of the equations of continuity and Eq. (4) is then solved numerically. This gives a new value for k. Its accuracy is tested against the previous one, the iterations being stopped when Eq. (4) reaches the value of 1 with a relative error (convergence criterion) that must be defined.

TABLE 1. Stiffness constants (in GPa) used for a Carbon fiber along *z* axis (3). The plane *xy* is isotropic.

c_{11}	c_{33}	c_{44}	c_{12}	c_{13}
19.82	234.74	24.0	9.78	6.36

Coupling Viscoelastic Losses and Multiple-Scattering

The theory proposed by Yang and Mal leads to a complex-valued wavenumber that describes both the phase velocity and the attenuation in the effective medium for the various polarities considered. In their results concerning Carbon-epoxy composite, both the matrix and the fiber materials were assumed to be purely elastic, that is to say, material properties were described by real-valued stiffness coefficients. The theory has not been applied directly in the case where the two materials (either one or both) are viscous, as it is the case for example in Carbon-epoxy composites.

To deal with viscoelastic components, we introduced in Yang and Mal theory complex-valued wavenumbers in both the matrix and the fiber appearing in the various computation stages of the unknown effective wavenumber.

The incident and scattered waves, wherever they propagate (matrix, fiber, effective medium), are thus described as waves subjected to attenuation. Attenuation symmetry in the various media is assumed to be identical to that of the elastic behavior. Therefore, all the stages of the computation of the original theory can be readily transformed by taking into account the complex-valued wavenumbers. By doing so, viscous losses are implicitly accounted for in the scattering process and both phenomena are coupled.

THEORETICAL RESULTS

Various results predicted with the proposed model are now given to illustrate its capabilities. In these results, composite materials made of Carbon fibers in an epoxy matrix with various fiber volume fractions are considered. Indeed, in the last part of the paper predicted attenuation coefficients will be compared with measured ones for such materials. Complex-valued Lamé's coefficients for the epoxy resin have already been given above. Resin density equals 1.23. That of a Carbon fiber equals 1.67. The stiffness constants used for the Carbon fibers are given as real-valued (elastic) constants given in Table 1.

The first results (Table 2) concern the stiffness constants one can deduce from the model since this model homogenizes the composite by transforming it into a homogeneous anisotropic attenuating medium. These results (computed for ϕ=65%) are deduced from the real part of k [Eq. (3)] at low frequency (k being frequency dependent). They are compared with others obtained by a classical static homogenization approach. Since only three cases of wave propagation (L, SV, SH) are computed, three constants can be deduced.

The full result concerning the variations of the real part of k with the frequency is shown on Fig. 3 for the same 65% case. Over a frequency range of [1 ; 10] MHz typical of frequencies used in UT, phase velocity (here for L waves) varies less than 0.02%. One can therefore conclude that in this case, phase velocity dispersion is negligible.

The next results show the variations of attenuation coefficient as a function of the frequency for various fiber volume fractions and as a function of the fiber volume fraction

TABLE 2. Stiffness constants (in GPa) for a Carbon-epoxy composite of 65% fiber volume fraction. Top: present model. Middle: classical approach. Bottom: relative error (%).

	c_{11}	c_{44}	c_{66}
Present model (GPa)	13.28	5.79	3.18
Classical approach (GPa)	13.22	5.77	3.12
Relative error (%)	0.5	0.3	1.8

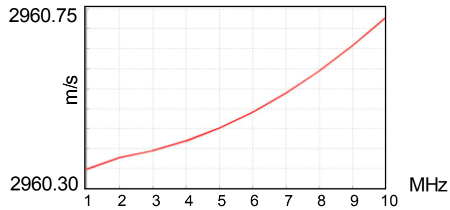

FIGURE 3. Variations of phase velocity with frequency for L waves propagating perpendicularly to fiber direction in an unidirectional layer with 65% fiber volume fraction.

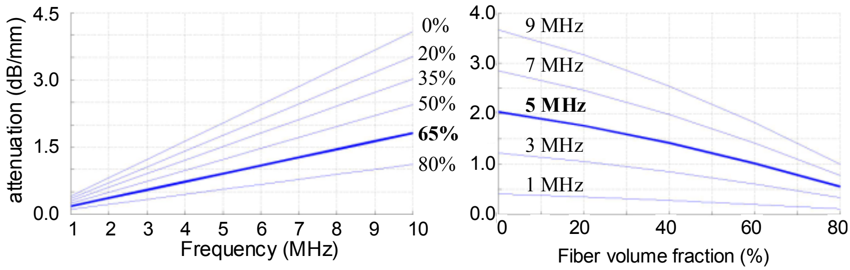

FIGURE 4. Variations of attenuation coefficient for L waves in a Carbon-epoxy composite. Left: *vs*. frequency for various fiber volume fractions. Right: *vs*. fiber volume fraction for various frequencies.

for various frequencies. Whatever the fiber volume fraction, attenuation increases linearly with frequency. Note that the model does not assume any specific law of attenuation (except a linear behavior for the pure epoxy). However, such a behavior (polynomial dependency of attenuation on frequency, of first degree in the present case) allows to model the attenuation of the Carbon-epoxy composite in the form given by Eq. (1).

This result is well-known by experimentalists though quite troublesome. Unfortunately, the model cannot help in interpreting this result further, since the physical meaning of computed quantities is a bit hidden by the iterative character of the algorithm.

Another interesting result shown here is that, for a given frequency, attenuation decreases as the fiber volume fraction increases. Viscoelastic losses due to the matrix lead to higher attenuation than multiple-scattering effects. Note that the same behavior is observed whatever the wave type (see last part of the paper for SH or SV waves).

In Fig. 5, attenuation coefficients predicted by the present model and by the model described in [4] as a function of the frequency are compared for Carbon-epoxy composites of two different fiber contents. At low (10%) fiber concentration, results from both models superimpose. At high (65%) concentration, the two models predict a lower attenuation but results are notably different (a difference of 0.7dB/mm at 5 MHz). Independent scattering approximation overestimates the attenuation which may appear as counterintuitive at first

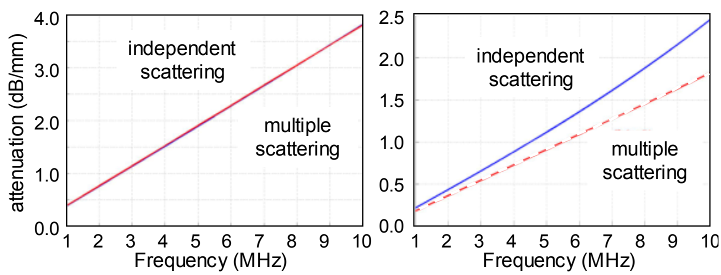

FIGURE 5. Predicted attenuation coefficients using independent scattering model and present multiple-scattering model. Left: $\phi = 10\%$. Right: $\phi = 65\%$.

glance. Under this latter approximation, part of the energy is definitely lost when back-scattered. If multiple-scattering is taken into account, some of this energy can be scattered again by other fibers and eventually propagate along the forward direction.

EXPERIMENTAL VALIDATION

Experimental measurements reported in Ref. [5] were compared to theoretical predictions. Measurements for the three possible polarities were made using wideband pulses, then processed at different frequencies. Various plates with various fiber volume fraction were manufactured. The manufacturing process is rather difficult and it was not possible to obtain a set of plates covering a large range of fiber volume fraction. The extreme case easily obtained was that of a plate made of pure resin (0% fiber volume fraction). Other plates were for fiber volume fractions varying in the range of [49 ; 61] %.

To illustrate the validation study, four plots comparing predicted and measured attenuation coefficients are shown on Fig. 6. The three polarities are considered (SH, SV, L), and for L waves, results at two different frequencies are shown.

Whatever the case, predicted attenuation coefficients are in excellent agreement with measured ones. These comparisons undoubtedly show the pertinence of the proposed model, at least in the case of Carbon-epoxy composite. For this composite, waves (incident and scattered) are attenuated by both viscoelastic losses in the matrix and multiple-scattering by fibers and therefore by their coupling.

Despite the lowest frequency (2 MHz) used in the SH case, the attenuation coefficient is the highest measured. In the cases dealing with L waves, one sees that the higher the frequency used in the measurement, the higher the attenuation. At the same frequency of 3 MHz, attenuation of SV waves is about three times higher than that of L waves.

CONCLUSION

A theoretical model has been developed to predict wave propagation and attenuation

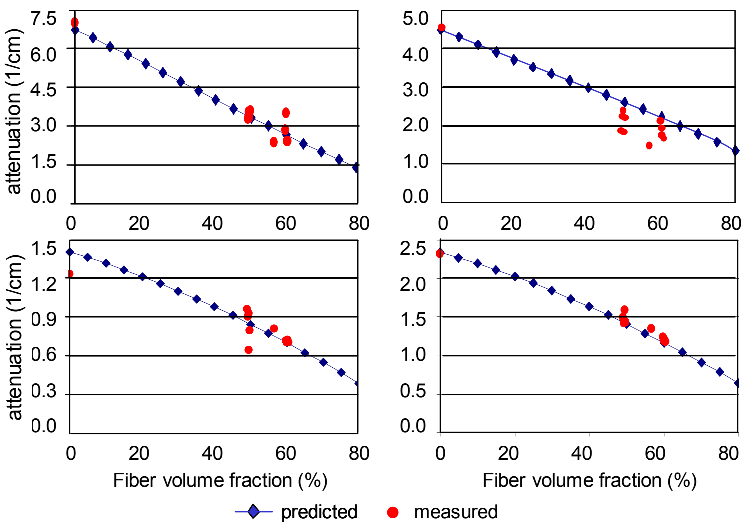

FIGURE 6. Predicted and measured [5] attenuation coefficients. Top left: SV wave at 3 MHz; Top right: SH wave at 2 MHz; Bottom left: L wave at 3 MHz; Bottom right: L wave at 5 MHz.

in unidirectional fiber reinforced composites. It couples effects due to multiple-scattering (as modeled by Yang and Mal theory) with viscoelastic losses by introducing complex-valued stiffness constants to describe the matrix (and possibly the fiber). The model is limited to the case of waves of arbitrary polarization propagating perpendicularly to the fiber direction. However, this corresponds to many cases of practical application in NDT.

At low fiber volume fraction, it was shown that predicted attenuation coefficients can be actually predicted without account of multiple scattering processes. At typical fiber volume fractions as those currently used for Carbon-epoxy composites (*e.g.* 65%), multiple-scattering plays an important role and must be accounted for.

In the case of Carbon-epoxy composites and whatever the fiber volume fraction, theoretical predictions show a linear frequency dependency of the exponential filter describing the attenuation law. Correlatively, phase velocity dispersion appears to be negligible.

Experimental measurements on Carbon-epoxy plates with various fiber volume fractions, at various frequencies and for various wave polarization relatively to the fiber direction were conducted. Excellent agreements of predicted attenuation coefficients with those measured validate the proposed model.

REFERENCES

1. Lhémery A., "Multiple-technique NDT simulations of realistic configurations at the French Atomic Energy Commission (CEA)", in *Review of Progress in QNDE,* Vol. 18, eds. D. O. Thompson and D. E. Chimenti, Kluwer Academic/Plenum Publishers, New-York, 1999, pp. 671-678.
2. Deschamps M. and Hosten B., "The effects of viscoelsaticity on the reflection and transmission of ultrasonic waves by an orthotropic plate", *J. Acoust. Soc. Am.* **91**, 2007-2015 (1992).
3. Varadan V. K., Ma Y. and Varadan V. V., "Multiple scattering of compressional and shear waves by fiber-reinforced composite materials", *J. Acoust. Soc. Am.* **80**, 333-339 (1986).
4. Biwa S., "Independent scattering and wave attenuation in viscoelastic composites", *Mech. Mater.* **33**, 635-647 (2001).
5. Biwa S., Watanabe Y. and Ohno N., "Analysis of wave attenuation in unidirectional viscoelastic composites by a differential scheme", *Comp. Sci. Tech.* **63**, 237-247 (2003).
6. Lonné S., Lhémery A. and Thévenot F., "A simulation study to explain the variability of ultrasonic attenuation measurement in RTM composites", in *Review of Progress in QNDE*, Vol. 23, eds. D. O. Thompson and D. E. Chimenti, AIP Conference Proceedings, Melville, New-York, 2004.
7. Biwa S., Idekoba S. and Ohno N., "Wave attenuation in particulate polymer composites: independent scattering/absorption analysis and comparison to measurements", *Mech. Mat.* **34**, 671-682 (2002).
8. Auld B. A., *Acoustic fields and waves in solids*, Wiley, N-Y, 1990.
9. Rokhlin S. I., Lewis D. K., Graff K. F. and Adler L., "Real-time study of frequency dependence of attenuation and velocity of ultrasonic waves during the curing reaction of epoxy resin", *J. Acoust. Soc. Am.* **79**, 1786-1793 (1986).
10. Yang R. B. and Mal A. K., "Multiple-scattering of elastic waves in a fiber-reinforced composite", *J. Mech. Phys. Solids* **45**, 1945-1968 (1994).
11. Waterman P. C. and Truell R., "Multiple-scattering of waves", *J. Math. Phys*. **2**, 512-537 (1961).