On the possible of the abnormally high damping effective properties of dispersion-reinforced composites and fibrous composites

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Abstract. We study the effective energy dissipation properties of solid polymer matrixes filled with coated spherical and fibrous inclusions coated with a layer of lossy viscoelastic material. The matrix and the inclusions are assumed to be separated by equal-thickness interfacial layers of a lossy viscoelastic material. We show that the remarkable loss amplification mechanism is operative in such particulate-morphology materials when the effective loss properties of composite can exceed the loss properties of the pure matrix by more than 20 times.

1. Introduction

In aerospace industry, the requirement for high vibrational damping and high strength/stiffness materials now seems to be mandatory since more structural parts of aircraft are being designed with reinforced polymer-matrix composite materials.

Several ‘traditional’ design optimization concepts and materials were proposed to enhance the damping properties of composites at the micromechanical or macromechanical level [1,2]. Here we shall concern ourselves with the way of optimizing the dynamic properties of composite material using viscoelastic layer, but the solution will be addressed from the nanotechnology perspective where an ultra thin coating layer of viscoelastic material is considered. With this kind of optimization, a remarkable high loss amplification effect can be observed. For a composite structure with inclusions approximately one hundred times stiffer than the matrix, the higher level of matrix damping can be obtained [3], i.e. the contribution of inclusions to damping can be very significant.

We elaborate a hybrid concept of viscoelastic and composite material to design composites with a good combination of high damping and high stiffness properties. The hybrid concepts are used by various authors to simultaneously obtain high damping and high stiffness composite using various analytical and computational methods [3-10]. Obtained results showed that there is a trade-off between its stiffness and damping properties.

In finding the optimal balance of properties in the work [11] examined the effect of thickness of viscoelastic coating layer surrounding spherical inclusions embedded in epoxy matrix system. It is found that at extremely thin coating layer of lossy material, the effective shear loss modulus of the composite increase substantially while the decrease in its effective elastic storage modulus is minimal. This high loss amplification effect is so extreme that its effective loss modulus value significantly exceeds the loss moduli of constituents in the composite. Similar results were reported later in [12] for
the wavy layered composites containing ultra thin viscoelastic layers. Based on the work [11] we can conclude that high damping and high stiffness composite structure might be attainable due to the presence of high shearing damping mechanism in lossy material (see also works [4,11,12]).

In this present work, we aim to examine analytically the effective dynamic properties (first approximation) of filled and layered composite material that has viscoelastic coating layer surrounding the inclusions, and also to understand their high damping mechanisms at the very thin layers of such lossy material. We show that the remarkable loss enhancement mechanism works in materials with particle morphology, when the effective loss properties of composite can exceed the loss modulus of the pure matrix by more than 20 times.

2. Filled composites reinforced by the spherical inclusions

To define the effective properties of the composite, we used self-consistent Eshelby’s method for the representative volume element (RVE) of spherical assembly, which consists of three concentric isotropic spheres embedded in an infinite effective medium with radiiuses \( R_1 < R_2 < R_3 \), the fourth layer representing the effective medium with infinite radius (\( R_4 = \infty \)).

The properties of spherical layered assembly are defined correspondingly by the bulk moduli of \( K_1 \) and \( \mu_1 \), spherical lossy layer with bulk modulus of \( K_2 \) and shear modulus \( \mu_2 \), moduli \( K_3 \) and \( \mu_3 \). The fourth layer representing the effective medium with infinite radius (\( R_4 = \infty \)) is characterized by unknown elastic properties of \( K_4 \) and \( \mu_4 \). According to the method, these are the effective elastic properties of the composite system where \( K_4 = K_{\text{eff}} \) and \( \mu_4 = \mu_{\text{eff}} \). Considered boundary problem and the problem of definition of the effective properties are solved exactly according to Eshelby’s procedure based on Christensen’s approach. Additional Eshelby’s equation was received in order to provide the necessary closure to the system of equations so that the effective properties of the composite material can be found. This additional equation is written as follow

\[
U' = \int_S (\sigma_{ij}^1 u_i^0 - \sigma_{ij}^0 u_i^1) dS = 0 \quad (i, j = 1, 2, 3),
\]

where \( U' \) is the increment of energy in a unit cell of matrix material containing the inclusion; \( S \) is the contact surface between matrix and effective medium; \( \sigma_{ij}^1, u_i^1 \) are taken to be the stress tensor and displacement vector components at the contact surface that are found from the contact problem; and \( \sigma_{ij}^0, u_i^0 \) are the stress tensor and displacement vector components on the contact surface, which are related to the conditions of the problem at infinity. Finally, we use correspondence principle approach to obtain the effective complex moduli of composite.

![Figure 1](image.png)

**Figure 1** Effective shear complex modulus of dispersed coated silica spheres in solid polymer composite with three different volume fractions (Continuous line – 50%, dashed line – 40%, dot-dashed line – 30%), Symbol \( \Delta \) represents the thickness of coating layer, \( \Delta = R_2 - R_1 \), and \( R \) is the radius of sphere \( R_1 \), volume fraction of inclusion, \( V = (R_2/R_3)^3 \).

As a result the displacements, stresses and effective properties were found using four bodies self-consistent Eshelby’s method and equation (1), [11,13]. The effective complex moduli of the
composite, were obtained (see figure 1) using viscoelastic correspondence principle proposed by Hashin.

The first phase was considered as an elastic material with \( \mu_1^* = b \), and the second phase is a polymer with \( \mu_2^* = a(1+i\eta) = \mu_2 + i\eta_\mu \). The effective complex moduli of the composite based on these properties are given in figure 1.

The following parameters were used for calculations: \( K_1 = 50 \) GPa, \( \mu_1 = 30 \) GPa, \( K_2 = 3 \) GPa, \( \mu_2 = 0.02 (1+i 0.5) \) and \( K_3 = 4 \) GPa, \( \mu_3 = 2.5 + i 0.005 \) GPa. The value of \( \eta \) is taken to be equal to 0.5 and the parameter for \( a \) is equal to 0.01 GPa, which are the typical values of polymer in the glass transition region. We can see very strong effect of increasing of the effective loss properties for considering composites. Upon varying the layers’ thickness, we detect two distinct peaks of energy dissipation, figure 1. The loss modulus at the second peak is 35 times larger than the loss modulus of the viscoelastic polymer-coating layer while at the first peak, the loss amplification effect is by a factor of 15 only.

Assume that composite is loaded by the homogeneous shear strain tensor at infinity and analyze deformations and the local dissipation energy in the lossy polymer layer. We relate the origins of the two peaks to two different states of local strain realizable inside the sections of maximum energy dissipation.

![Figure 2](distribution_of_the_local_strain_tensors_in_lossy_layer_at_thickness_where_the_maximum_dissipation_energy_of_peaks_occurs_for_different_relative_thickness_of_lossy_layer.png)

**Figure. 2** Distribution of the local strain tensors in lossy layer at thickness where the maximum dissipation energy of peaks occurs for different relative thickness of lossy layer.

We can see from figure 2 that the strain magnification factor at second peak is substantially higher. Interestingly, the second peak behavior also corresponds to both maximum dissipation energy and maximum total average energy density. The first peak where \( \Delta/R \approx 0.2 \) the strain magnification factor is \( \gamma_1 \approx 5 \) and at the second where \( \Delta/R \approx 0.005 \), a factor of \( \gamma_2 = 45 \). Note that for the first peak at \( \Delta/R = 0.2 \) the Energy dissipation rate is 0.198789 and for the second peak the energy dissipation rate is 21.6935. So, it is obvious now that the high damping capacity of the composite is attributed to the presence of unusual high shearing damping mechanism in the lossy layer.

Based on the analytical estimates for the two different effective complex moduli in two different system of composite material, we observe that the appearance of very high shear loss amplification effect is due to the presence of an ultra thin coating layer of viscoelastic material in the composite. At that optimal thickness of coating layer, the effective loss moduli increases substantially and it exceeds the loss moduli of viscoelastic and matrix material significantly. Interestingly, at ultra thin coating of viscoelastic layer of second peak, the corresponding volume fraction is approximately 0.1%. On the other hand it can be shown that the role of normal strain damping mechanism in the lossy layer in
optimizing the damping capacity of such structure is minimal. Note that increasing the volume fraction of inclusion increases the loss and storage characteristics of the composites.

3. **High loss amplification effect in lamellar media**

Let us consider now the simple problem of lamella-type composite loaded separately by shear stresses and or normal transversal stresses.

The effective complex shear and transverse Young’s moduli can be determined using Reuss estimation with viscoelastic correspondence principle. Thus, their effective complex moduli can be written as follows: $1/\mu_{eff} = (1-V)/\mu_1 + V/\mu_2$, $1/E_{eff} = (1-V)/E_1 + V/E_2$; where $V$ is the volume fraction of second phase; $\mu_1$ and $\mu_2$ are the complex shear moduli of the first and second phase respectively; $E_1$ and $E_2$ are their transverse Young’s moduli respectively. For shear case, we consider the first phase as an elastic material with $\mu_1 = b$, and the second phase is a polymeric material with $\mu_2 = a(1+i\eta) = \mu_2 + i\mu_2'$. Let us study behavior of the considered composite material under external harmonic strain $\varepsilon = \varepsilon_0 \sin(\omega t + \delta)$, in the infinity with a given angular frequency $\omega$, $\varepsilon_0$ is the amplitude of harmonic strain applied to a viscoelastic continuum. Then in the steady-state the system stress is also harmonic $\sigma = \sigma_0 \sin(\omega t + \delta)$. At specified cyclic frequency, the typical complex modulus is defined by the real part represents the storage modulus and the imaginary part represents the loss modulus of material. The imaginary part is also responsible to formally define the rate of energy dissipation per unit volume as $W = E''E'$. The ratio of $E''/E'$ is called loss tangent or loss angle (tan $\eta$), $\tan \delta = E''/E'$ which is related to the damping of a material, and $\eta$ is the out-of-phase angle between harmonic stress and strain in sinusoidal loading. Figure 3 demonstrates the benefit of viscoelastic polymers (at $T_g$) when compared to solid polymer (below $T_g$) for the following parameters of composite: the shear modulus of elastic phase, $b = 30$ GPa, solid polymer (below $T_g$), $a_m = 1$ GPa and $\eta_m = 0.02$, and viscous polymer (at $T_g$), $a_v = 0.01$ GPa and $\eta_v = 1$. The simple analytical estimations show that the effective shear loss modulus at a very thin layer of viscoelastic material significantly exceeds the effective shear loss modulus of solid polymer-matrix ($\eta_m = 0.02$) almost by 300 times and its solid polymer composite by 30 times. It is easy to show that even for the epoxy matrix with very small damping properties $\eta = 0.005$, the maximum of the effective shear loss modulus of composite realizes for thin enough layer of epoxy matrix $V=0.1$ and exceeds the effective shear loss modulus of epoxy matrix ($\eta_m = 0.005$) almost by 5 times (see figure 3 (a)). On the other hand using the solid polymer (below $T_g$), and epoxy matrix lead to composites with higher effective storage modulus (see figure 3 (b)).

![Figure 3](image_url)

**Figure 3.** Effective shear complex modulus of lamella composite with different types of polymers; (a) - Loss modulus (dotted line -at $T_g$, dashed line - below $T_g$); (b) - storage modulus (dotted line-at $T_g$, dashed line - below $T_g$, solid line-epoxy matrix).
Note that for the bulk modulus there is a similar dependence, but with a different magnification factor at the maximum. It is important that for the materials under consideration, the magnification factor significantly lower in comparison with shear modulus, and the maximum is realized at higher than for shear volumetric contents of a viscous polymer. We can conclude that the abnormally high effective damping properties of a composite with simultaneously high effective mechanical properties are realized for the second peak corresponding to the shear mode.

It is important to note also that for a layered system, the shear mode and bulk mode can be considered separately, while in the general case these modes are realized jointly. Therefore, in the general case, there are two peaks, as shown in figure 1.

In this regard very useful can be to following statement.

Statement. Let us consider a composite with inclusions containing viscoelastic coatings. To estimate the optimal thickness of the viscoelastic layer providing the maximum effective damping properties of the composite, it is enough to consider a lamellar media with the same volume content of a viscoelastic polymer and find the volume content of a viscoelastic polymer corresponding to the maximum effective shear loss modulus. There is an analytical estimation of this value, which provides the optimal value of the thickness of the viscoelastic coating; \( \Delta R \approx V = (a/b) \sqrt{(1 + \eta^2)} \). Direct numerical estimates show that this statement is also true for fibrous composites with fibers containing viscoelastic coatings.

4. Composites with cylindrical fibrous inclusions containing viscoelastic coating layers

The dissipation mechanisms discussed above are fully transferred to fibrous composites with fibers having viscoelastic coatings.

We consider a unidirectional fiber composite lamina, which is a transversely isotropic material, and designate axis 3 as axis of symmetry parallel to the principle axis of fiber (z-axis). As before, self-consistent Eshelby’s method is used \([14,15]\) to examine all five necessary effective properties, which are required to fully describe the behavior of transversely isotropic lamina. These effective properties are plane strain bulk modulus \( K_{11}^{\text{eff}} \), axial Young’s modulus \( E_{33}^{\text{eff}} \), axial \( \mu_{23}^{\text{eff}} \) and transverse shear modulus \( \mu_{12}^{\text{eff}} \), and Poisson’s ratio \( \nu_{31}^{\text{eff}} \). From these effective properties, transverse Young’s modulus can be obtained.

![Figure 4](image-url)

**Figure 4.** The dependence of effective transversal shear complex modulus (a) and axial shear complex modulus (b) of unidirectional composite lamina on the coating thickness of viscoelastic polymer. The continuous line represents 50%, dashed line represents 40%, and dot-dashed line represents 30% volume fraction of coated inclusions. Symbol \( \Delta \) represents thickness of coating layer \( (\Delta = R_2 - R_1) \) and \( R \) is the radius of fiber, bulk moduli for glass fiber for polymer coating and for Epoxy matrix are \( K = 50 \) GPa, \( K = 3 \) GPa and \( K = 2.5 \) GPa; shear moduli for fibres, polymer and for epoxy matrix are respectively \( \mu = 30 \) GPa, \( \mu = 0.02 + i \) 0.01, \( \mu = 2.5 + i \) 0.005 GPa.
In the general case, as a result of the analysis of the stress state that is introduced to determine the effective properties, it can be argued that only for the transverse shear two dissipation modes are realized simultaneously - the shear and the bulk modes. Here we can see two peaks for loss modulus (see figure 2 (a)). In other cases, only one peak has place (see as example figure 2 (b)) and estimates of the loss modulus can be done directly on the basis of layered structures, where these modes are implemented separately.

5. Conclusion
The present paper presents a model explaining the emergence of non-traditional high-strength damping properties of composite materials containing inclusions with layers of viscoelastic coating in the range of small coating thicknesses. It is shown that to determine the optimal thickness of a viscoelastic layer, one can use the analytical estimate obtained for lamellar media.

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