High Damping Composite Materials: Effect of Structural Hierarchy

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ABSTRACT: A combination of stiffness and loss (the product $E\tan\delta$) is desirable in damping layer and structural damping applications. Composite materials of structure which gives rise to Reuss or Hashin–Shtrikman lower bound behavior can give rise to such properties. Hierarchical particulate morphologies attain the Hashin–Shtrikman curve. We show that hierarchical composites give rise to complex Poisson's ratios which, however, have minimal effect on the stiffness-map. We show that structural hierarchy is useful in viscoelastic composites in that it enables the attainment of high concentrations of spherical inclusions, and that it facilitates the attainment of both stiffness and damping. A damping layer upon a substrate is considered as the top level of the structural hierarchy. We demonstrate that if the layer itself is a relatively stiff composite, the penalty usually associated with such a geometry for compliant layers is ameliorated.

INTRODUCTION

ENERGY ABSORPTION BY mechanical damping is of considerable importance in that one can damp vibrations in mechanical systems; high damping materials are used to reduce vibration in aircraft and other machinery. The benefit is longer service life of components, reductions in weight, and reductions in noise. The loss tangent $\tan\delta = \mathrm{Im}(E^*)/\mathrm{Re}(E^*)$ as a measure of damping is the ratio of the imaginary part to the real part of the complex modulus E^*. The angle δ is the phase angle between stress and strain sinusoids. Tan δ is proportional to the energy loss per cycle within the framework of linear viscoelasticity.

The product $|E^*|\tan\delta$, is a figure of merit for materials used as damping layers. Figure 1 shows a stiffness-loss map, $|E^*|$ vs. $\tan\delta$, of common materials. Structural materials

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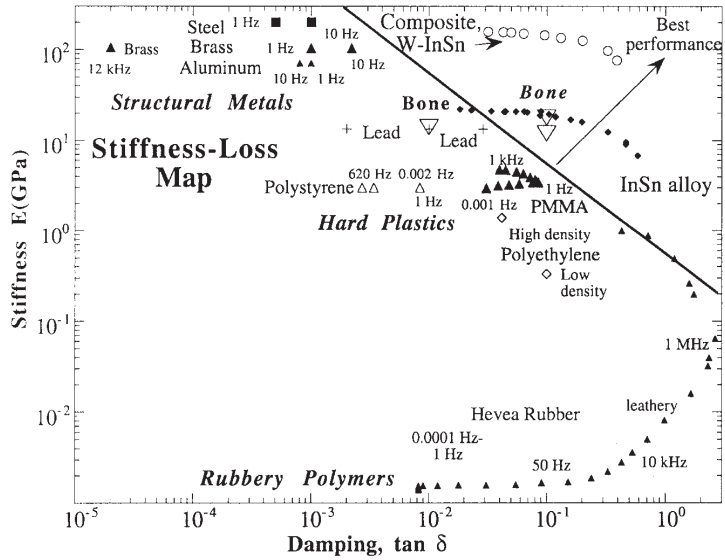

Figure 1. Stiffness-loss map of observed behavior of some common materials, and designed high-loss materials, adapted from Lakes [2,23].

occupy positions to the left or far to the left in the diagram, i.e., low-damping, high modulus. Rubbery materials appear in the lower right. Materials that combine high damping and high stiffness are not common.

Several kinds of composite micro-structure give rise to high stiffness combined with high viscoelastic loss. Two phase composites composed of a stiff, low loss phase and a compliant high loss phase can exhibit high stiffness combined with high loss tangent [1] provided the structure is that of a Reuss laminate, a material containing soft platelets, or a material describable by the Hashin–Shtrikman "lower" formula. The Hashin–Shtrikman formulae provide upper and lower bounds for stiffness vs. volume fraction of elastic isotropic composites. The Hashin–Shtrikman "lower" curve appears in the upper right on a stiffness-loss map, and is close to the Reuss curve, $1/E_c^* = V_1/E_1^* + V_2/E_2^*$. Therefore anisotropy in viscoelastic composites with arbitrary volume fraction differs in its effects from elastic composites with fixed volume fraction. Anisotropy does not significantly expand the stiffness loss map, even for the favorable case of a shear load. By contrast, in the usual plots of stiffness vs. volume fraction for elastic composites, a substantial gain in stiffness for a given volume fraction can be achieved by introducing anisotropy. Favorable viscoelastic properties are most easily achieved if the stiff phase is as stiff as possible [2]. Inclusion of a small amount of damping in the stiff phase has little effect on the composite damping [2]. The bounds [3] for viscoelasticity in the stiffness-loss map either coincide with the Hashin–Shtrikman formulae or are close to them [4,5]. Structures which attain the Hashin–Shtrikman formulae are shown in Figure 2.

Composites based on Reuss or Hashin–Shtrikman "lower" morphologies give high stiffness and damping. The compliant, high damping phase is continuous in either case. Polymers or soft metals could be used for the high damping phase. Since such materials are usually weak (have a low yield strength) compared with structural metals, such composite will also be weak. That is not a problem since the high damping composite can be used as a damping layer upon a structural material. Reuss laminates were used in an

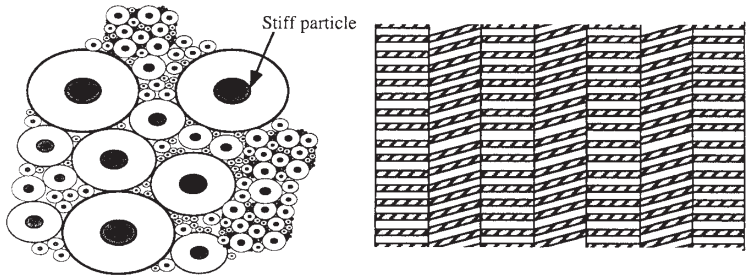

Figure 2. *Extremal hierarchical composite morphologies: (a) coated spheres morphology and (b) rank two laminate morphology.*

experimental demonstration (Figure 1) of high-loss stiff materials [2]. Tungsten, with a Young's modulus twice that of steel, was used as the stiff phase. Indium-tin alloy was used as the high damping phase. In view of the cost and density of tungsten, alternate stiffness phases such as silicon carbide, available in particle form, are under study in our laboratory.

In most prior analyses of viscoelastic composites, the emphasis has been upon determining properties of structures already decided upon for reasons other than viscoelasticity. In this article we are concerned with optimal structures to maximize damping. In particular, we consider the effect of structural hierarchy in simultaneously achieving high stiffness and high damping, with emphasis upon particulate morphologies which are amenable to laboratory fabrication at moderate cost.

ANALYSIS OF HIERARCHICAL COMPOSITES

For phases assumed to be linearly viscoelastic, the effective dynamic functions of the composite were calculated from constituent properties and known solutions for elastic heterogeneous media by the dynamic correspondence principle of the theory of linear viscoelasticity [6,7]. A solution for an elastic problem is taken as a starting point, and each elastic modulus is replaced with a complex dynamic modulus in the analysis. Real and imaginary parts are separated, and the absolute value of the complex modulus is plotted vs. $\tan\delta$ to produce a stiffness loss map.

For elastic composites, the lower bond for the shear modulus G_L of the composite is given by the Hashin–Shtrikman relation [8]

$$G_L = G_2 + \frac{V_1}{1/(G_1 - G_2) + (6(K_2 + 2G_2)V_2)/(5(3K_2 + 4G_2)G_2)} \tag{1}$$

in which K_1, K_2, G_1, G_2, V_1 and V_2 are the bulk modulus, shear modulus and volume fraction of phases 1 and 2, respectively. Here $G_1 > G_2$, so that G_L represents the lower bound on the shear modulus. Interchanging the numbers 1 and 2 in Equation (1) gives the upper bound G_U for the shear modulus.

Composites obeying the Hashin–Shtrikman formulae are attained via certain hierarchical microstructures. For example the coated spheres structure [Figure 2(a)] attains the Hashin–Shtrikman bounds for bulk modulus, and the rank-two laminate structure [9] [Figure 2(b)] attains the above Hashin–Shtrikman bounds for shear modulus [10,11].

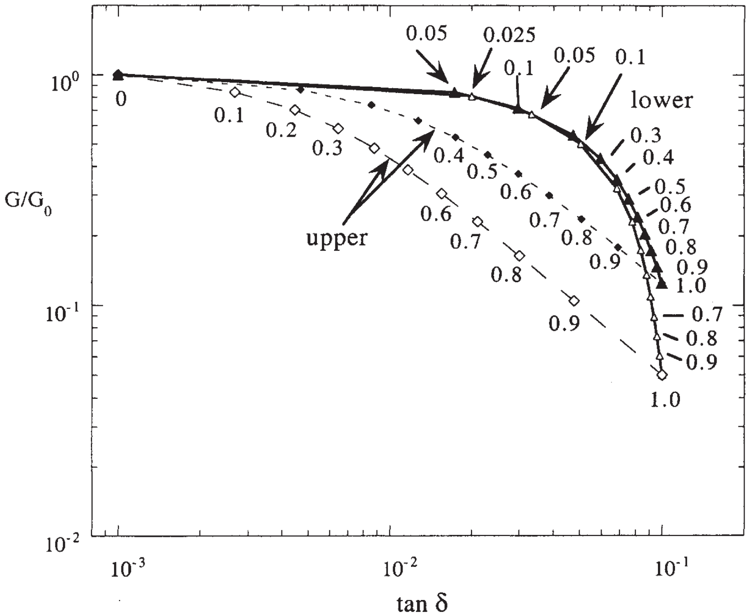

Figure 3. Stiffness-loss map for different soft phase stiffness for Hashin–Shtrikman composites with phase Poisson's ratio 0.3. Top curves, inclusion/matrix modulus ratio 8; bottom curves, modulus ratio 20. Volume fraction increments of 0.1, as shown by numbers within the frame, with extra increments for the "lower" formula at small volume fraction of the high-damping phase. Upper and lower refer to the corresponding Hashin–Shtrikman formulae which are upper and lower bounds for elastic modulus for given volume fraction. G_0 is the stiffness of the stiff phase. For a comparatively stiff high-damping phase, a smaller volume fraction of stiff inclusions suffices to maximize the product of composite stiffness and damping.

For viscoelastic materials, apply the correspondence principle to Equation (1). The complex viscoelastic shear modulus G_L^* of the composite via the "lower" formula is:

$$G_L^* = G_2^* + \frac{V_1}{1/(G_1^* - G_2^*) + (6(K_2^* + 2G_2^*)V_2)/(5(3K_2^* + 4G_2^*)G_2^*)} \tag{2}$$

The "upper" formula for G_U^* is obtained as above. In the stiffness-loss maps in Figure 3 the effect of the stiffness of the soft phase for a Hashin–Shtrikman composite is illustrated. Numbers within the frame refer to volume fractions. Curves are normalized to G_0 which is the stiffness of the stiff phase. The curves in the upper right region are derived from the Hashin–Shtrikman "lower" formula. These curves for different stiffness values of the soft phase are similar in shape. However if the high-damping phase is made stiffer, a smaller volume fraction of stiff inclusions is required to achieve high stiffness and high damping. This would be inconsequential in a laminate, since it is not difficult to make laminae of different thickness. We remark that the laminate Reuss formula gives rise to a similar curve shape as the "lower" Hashin–Shtrikman formula in the stiffness loss map [2]. However use of a stiffer damping phase simplifies a particulate embodiment since in practice it can be difficult to achieve a high volume fraction of particles. As an example of the modulus ratio 20 in Figure 3, the inclusions may be tungsten or silicon carbide, with a Young's modulus of about 400 GPa, and the matrix indium-tin alloy, with a Young's modulus of about 20 GPa and high damping. For a modulus ratio of 8, the matrix could be tin, with a Young's modulus of about 50 GPa. Pure cast tin has a $\tan\delta$ of only about 0.02 to 0.03, however damping can be increased to at least 0.06 by permanent deformation [12]. Zinc as a matrix has a Young's modulus $E = 100$ GPa, giving a modulus ratio of 4 in these composites, and $\tan\delta = 0.01$ to 0.02 for cast zinc and up to 0.04 for permanently deformed zinc. Study is in progress to further increase the damping of matrix materials.

Consider for comparison hierarchical composites containing spherical inclusions. For a small volume fraction $V_{so} = 1 - V_{st}$ of spherical elastic inclusions in a continuous phase of another elastic material, the shear modulus of the composite G_{i+1} is given by the following relation [13]. The expression is written recursively to allow hierarchical structure; $i+1$ represents the level of structural hierarchy. At the lowest level, $i = 0$, and $V_i = V_{so}$.

$$\frac{G_{i+1}}{G_i} = 1 - \frac{15(1-\nu_i)(1-(G_{st}/G_i))V_{st}}{7 - 5\nu_i + 2(4 - 5\nu_i)(G_{st}/G_i)} \tag{3}$$

in which ν_i is the Poisson's ratio of phase i (the matrix, here considered softer than the inclusions); phase st represents the (stiff) inclusion material. The soft phase itself is considered hierarchical, hence the subscript i. We assume the Poisson's ratio is 0.3.

High concentrations of particles may be achieved by a hierarchical scheme shown in Figure 4. Rank one refers to a dilute suspension of spherical inclusions as shown on the left. In rank two, the matrix itself contain spherical inclusions, of sufficiently small size that it may be regarded as a continuum as illustrated schematically in idealized form on the right. For a rank two composite, Equation (3) is applied once recursively; for rank three it is applied twice.

For viscoelastic composites, the correspondence principle is again invoked, and elastic constants replaced by complex quantities signifying viscoelastic behavior. In Figure 5, the Hashin–Shtrikman "lower" composite is compared with the hierarchical composite with spherical inclusions. As in Figure 3, curves are normalized to G_0 which is the stiffness of the stiff phase. Observe there is little difference between these composites in the stiffness-loss map. Structures which attain the Hashin–Shtrikman formulae (coated spheres and hierarchical laminate) are already hierarchical, so the comparison is of one hierarchical material with another.

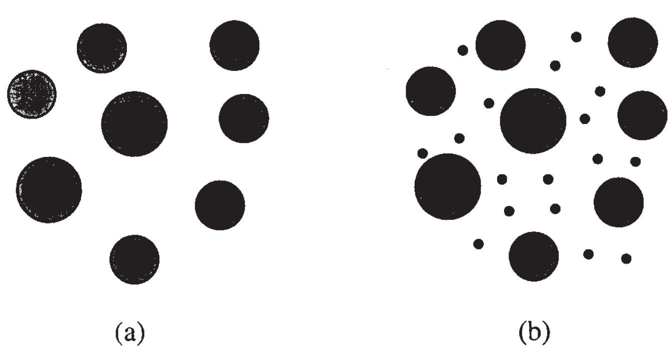

Figure 4. Hierarchical spheres morphology: (a) rank one, spherical inclusions and (b) rank two. The matrix itself contains spherical inclusions, of sufficiently small size that it may be regarded as a continuum.

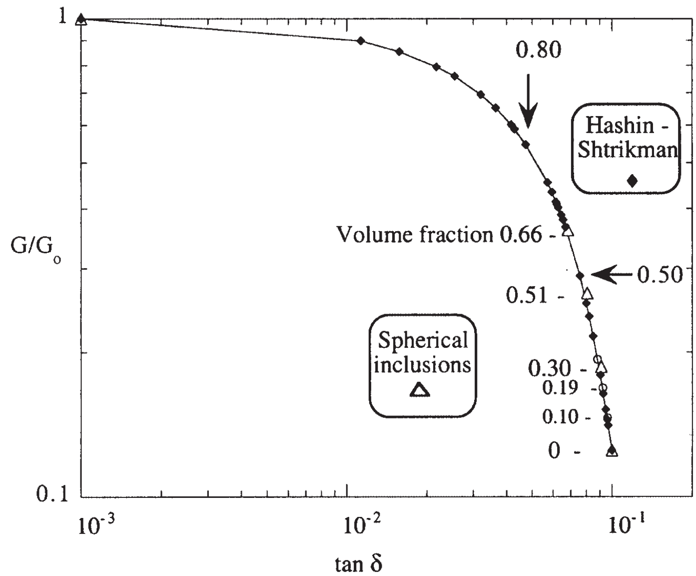

Figure 5. *Stiffness-loss map of calculated properties of hierarchical composite of spherical inclusions, compared with Hashin–Shtrikman "lower" composite.*

Phase Angles in Poisson's Ratio

Poisson's ratio ν appears indirectly in the Hashin–Shtrikman formulae for shear modulus G and bulk modulus K since for isotropic materials $\nu=(3K-2G)/(6K+2G)$ and $K=2G(1+\nu)/3(1-2\nu)$. Application of the correspondence principle allows each mechanical property term to be complex in a viscoelastic material. It is therefore appropriate to consider the effect of complex Poisson's ratios. Hashin–Shtrikman composites with phases with real Poisson's ratios give rise to a composite Poisson's ratio which is complex, as shown in Figure 6. For typical values of Poisson's ratio for stiff constituents, the phase angle in Poisson's ratio is considerably less than the viscoelastic phase angle δ in the compliant portion. Phase angles in Poisson's ratio can influence the bounds [5]. Suppose the matrix is itself a composite with a relatively small phase angle in Poisson's ratio. There is as a result little influence on the stiffness loss map of the composite as shown in Figure 7. Indeed [5], the loss tangent associated with the bulk modulus of the composite can be no larger than the maximal and no smaller than the minimal loss tangent (for the bulk of shear deformations) of either phase. Since the structures considered here give rise to behavior approaching or attaining the bounds, it is not surprising that phase angles in Poisson's ratio do not give rise to dramatic changes in the damping.

Stiff Damping Layers

In practical vibration absorption systems, a layer of damping material, commonly a polymer, is attached to a structural material, commonly a metal or fibrous composite. To use a hierarchical composite damping material, the layer-substrate structure represents

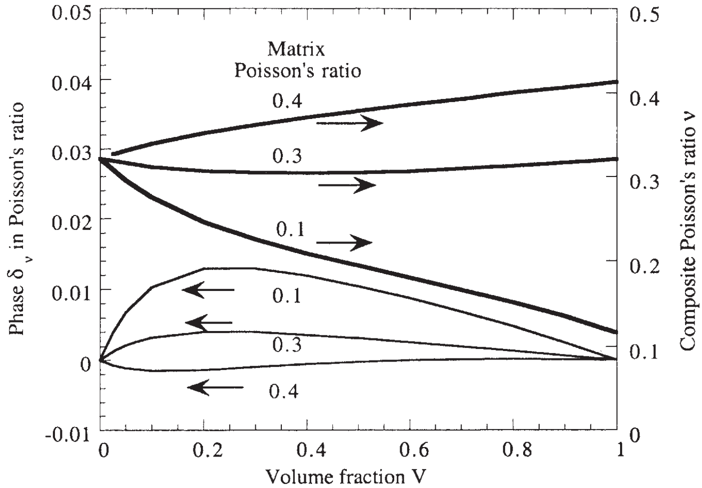

Figure 6. Poisson's ratio and phase angle ν of Poisson's ratio in Hashin–Shtrikman "lower" composite, for a modulus ratio of 8, an inclusion Poisson's ratio of 0.33, and for several real values of matrix Poisson's ratio. The compliant phase had $\tan\delta = 0.1$, the stiff phase 0.001.

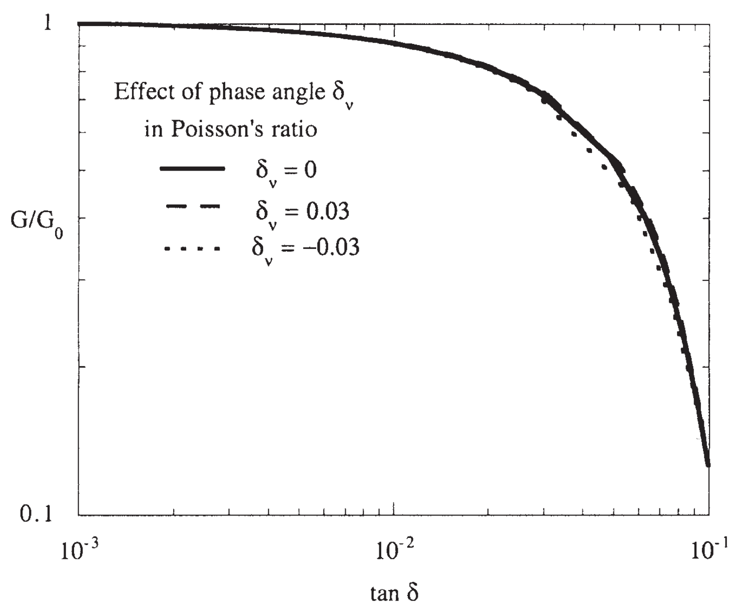

Figure 7. Stiffness-loss map showing that a moderate phase angle in the Poisson's ratio has a minimal effect. The matrix $\tan\delta = 0.1$. Phase angle in Poisson's ratio: 0, solid line; 0.03, dash line; -0.03, short dash line.

the topmost level of the structural hierarchy. Ordinary damping layers made of polymeric materials are orders of magnitude more compliant than their substrate. The layered system considered as a composite is Voigt, $E_c^* = E_1^* V_1 + E_2^* V_2$, if it is in tension/compression. The effect of the layer is expressed in terms of the damping $\tan\delta_d$ of the layer as follows.

For a thin layer of loss modulus E''_d and thickness t_d, upon a stiff plate [14] which has storage modulus E'_p and thickness t_p,

$$\tan\delta_{eff} \approx \frac{E'_d \tan\delta_d}{E'_p}\frac{t_d}{t_p} \tag{4}$$

The effect of a thin damping layer is also voigt-like if, as is usual, it is in bending. It is easier to achieve effective damping in bending, as follows, than in tension/compression or shear.

$$\tan\delta_{eff} \approx 3\,\frac{E'_d \tan\delta_d}{E'_p}\frac{t_d}{t_p} \tag{5}$$

For polymer layers upon metal, $E'_d \ll E'_p$, so the effective system damping is usually much less than the damping of the layer. If, however, a damping layer is made of a composite of a high concentration of ceramic inclusions ($E \approx 400$ GPa) and a metal of low melting point ($E \approx 50$ GPa), the layer may be as stiff as or even stiffer than common structural substrates such as aluminum ($E = 70$ GPa) or steel ($E = 200$ GPa). As shown in Figure 8, for damping layers with a stiffness comparable to that of the substrate, the penalty associated with Voigt or Hashin–Shtrikman "upper" morphology is minimal. Arrows show 50% volume

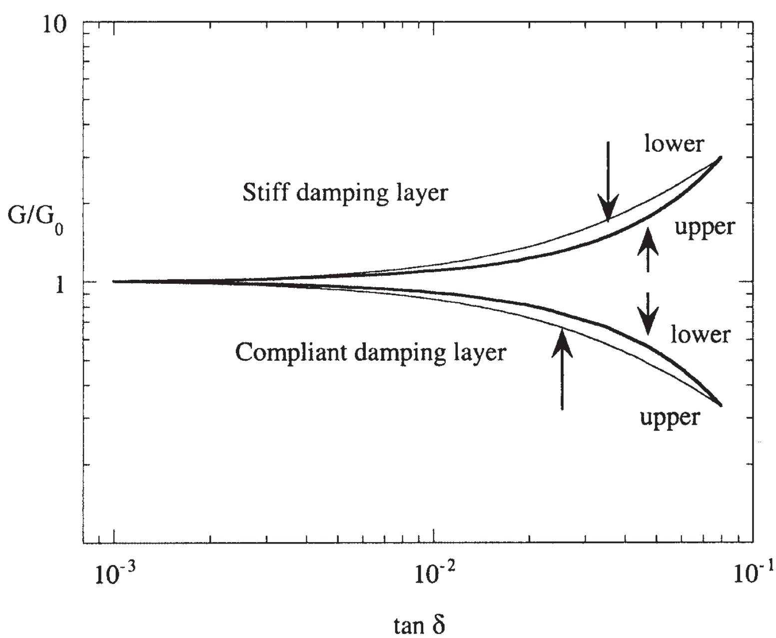

Figure 8. *Stiffness-loss map for upper and lower Hashin–Shtrikman composites, assuming the lossy phase is one-third as stiff, or three times stiffer than the low-loss phase. For the layer, $\tan\delta = 0.08$; for the substrate, $\tan\delta = 0.001$. Upper and lower refer to the corresponding Hashin–Shtrikman formulae which are upper and lower bounds for elastic modulus for given volume fraction. For stiff damping layers, the penalty associated with Voigt or "upper" morphology is minimal. Arrows show volume fraction 0.50. Such stiff, lossy phases are possible if they are themselves composites.*

fraction. As a further example consider a Voigt laminate of a layer of steel ($\tan \delta = 0.001$) with a damping layer of equal thickness. If the damping layer is rubbery with $E = 10$ MPa and $\tan \delta = 1.0$, then the layered structure is not significantly stiffened and its effective $\tan \delta = 0.00104$. The damping layer is therefore ineffective in tension/compression. A polymeric layer in the leathery regime, with $E = 100$ MPa and $\tan \delta = 1.0$, gives an effective structural $\tan \delta = 0.0015$, still not much more than the steel itself. A composite damping layer with $E = 100$ GPa and $\tan \delta = 0.05$, gives an effective structural $\tan \delta = 0.017$, a notable improvement. Such layer properties are attainable using the composite principles articulated here and elsewhere. Moreover the stiff layer gives a 50% increase in structural stiffness in this example.

Exceeding the Bounds

All bounding calculations depend on certain assumptions. Specifically, the bounds for elastic and viscoelastic behavior referred to above assume linear isotropic behavior, and that the phases are perfectly bonded. It is tacitly assumed that only deformation fields carry an energy density. If the last assumption is relaxed, one can exceed the bounds. In materials describable by coupled fields, the other field variables can have an associated energy density. One may, of course, derive new bounds which incorporate the additional freedom of coupled fields.

For example, in thermoelasticity, there is a coupling between mechanical variables and thermal variables such as temperature and entropy. The strength of the coupling depends upon the thermal expansion coefficient, therefore the coupling is almost always present. In a composite, a suddenly applied load gives rise to a heterogeneous deformation field, hence a heterogeneous temperature field. Heat flow in response to stress [15,16] can give rise to mechanical damping in composites even if each phase is elastic under isothermal conditions. The maximum $\tan \delta$ attainable in known composites by thermoelastic coupling is about 0.01 [17].

Piezoelectricity is also a coupled field effect. In piezoelectric materials such as quartz, Rochelle salt, and lead titanate zirconate ceramics, stress and strain are coupled to electrical field and polarization. Only those materials lacking a center of symmetry on the atomic scale can be piezoelectric. Electrical conductivity, external circuits [18], or phase angles in the dielectric or piezoelectric properties [19] can give rise to mechanical loss via electromechanical coupling. The maximum $\tan \delta$ attainable can be large, greater than 0.5. As with thermoelastic materials, piezoelectric materials as composite phases can give rise to larger damping than anticipated based solely on mechanical considerations.

Magnetostriction is a coupled field effect which, as with piezoelectricity, finds application is transducers. Certain magnetostrictive composites give rise to large response [20].

DISCUSSION AND CONCLUSION

High-loss composites are of interest for the following reasons. In current technology, polymer layers are able to suppress bending vibration of thin plates. They have the disadvantages of temperature sensitivity, flammability, a narrow effective range of temperature and frequency, and a comparative inability to control vibration in deformation modes other than bending. For free layer damping, the figure of merit for the layer material is the product of Young's modulus and mechanical damping: $E\tan\delta$. Therefore a combination of high stiffness and high damping is beneficial, but difficult to attain in polymers. For polymers of the highest damping ($\tan\delta > 1$), the full width at half maximum of the damping peak at constant frequency may be only about 18°C [21]. This is adequate for machinery which operates near room temperature, but is insufficient for aircraft and automobiles in which the skin temperature may vary over a considerable range. Composites based on a high-loss metal matrix can give damping over a wide range of frequency, so they are likely to be promising over a wide range of temperature. A damping layer which is sufficiently stiff will have the further effect of stiffening the structure, raising the natural frequencies to values less easily excited.

As for structural hierarchy, a simple rank one Reuss laminate is described by a stiffness-loss map similar to that of a Hashin–Shtrikman "lower" formulation, which is attainable by a hierarchical structure. Therefore structural hierarchy is not necessary to achieve stiff, high loss composites. This is in contrast to the problem of compressive strength of cellular solids, in which structural hierarchy can give rise to orders of magnitude improvement [22]. Complex Poisson's ratio occurs in hierarchical composites, however for the cases considered, the effect on the stiffness-loss map is negligible. Structural hierarchy is nonetheless useful in viscoelastic composites in that it enables the attainment of high concentrations of spherical inclusions. Damping layers made of such composites upon a stiff, strong, but low damping substrate allow the attainment of stiffness, damping, and strength.

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