Analysis of high loss viscoelastic composites

by

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Synopsis

A theoretical study of viscoelastic properties of composites is presented with the aim of identifying structures which give rise to a combination of high stiffness and high loss tangent. Laminates with Voigt and Reuss structure, as well as composite materials attaining the Hashin-Shtrikman bounds on stiffness were evaluated via the correspondence principle. Similarly, viscoelastic properties of composites containing spherical or platelet inclusions were explored. Reuss laminates and platelet filled materials composed of a stiff, low loss phase and a compliant high loss phase were found to exhibit high stiffness combined with high loss tangent.

INTRODUCTION

Viscoelastic materials can be of use in the damping of mechanical vibration and in the absorption of sound. The loss tangent, or tangent of the phase angle δ between stress and strain in sinusoidal loading, is a useful measure of material damping. Most materials used in structural applications, however, have small loss tangents. Conversely, materials with high loss tangents tend to be compliant, hence not of structural interest. Fig. 1 contains a stiffness-loss map (plot of the absolute value of the dynamic modulus vs loss tangent) for some representative materials. Compliant, lossy materials are used as layers over stiff materials in various applications; nevertheless a stiff material with high loss would be of use in structural damping of noise and vibration. We consider in this article the possibility of making composite microstructures providing high stiffness and high loss.

A possible avenue for making high loss composites is to make use of non-affine deformation. This is in contrast to affine deformation in which the particles in the solid move in a way corresponding to a uniform strain plus a rotation in a continuum. The negative Poisson's ratio materials developed by Lakes (1987) exhibit this property in that the foam cells unfold during deformation (Lakes, 1991; Chen and Lakes, 1991). Non-affine deformation can result in high viscoelastic loss in a composite if the phase which has the highest loss experiences a larger strain than does the composite as a whole.

Elastic properties of multi-phase composite materials have been studied extensively. Of these studies, the ones most relevant to the present work are those dealing with bounds on the elastic behavior and predicted properties of composites of relatively simple structure. The upper and lower bounds of stiffness of two phase and many phase composite materials have been obtained in terms of volume fraction of constituents (Hashin, 1962; Hashin and Shtrikman, 1963). Bounds and expressions for the effective elastic moduli of materials reinforced by parallel hollow circular fibers in hexagonal or random arrays have also been derived by a variational method (Hashin, 1962). Furthermore, bounds on three independent effective elastic moduli of an n-phase fiber reinforced composite of arbitrary transverse phase geometry, plane strain bulk modulus, transverse shear modulus and shear modulus in plane parallel to fibers, have been derived in terms of phase volume fractions(Hashin, 1965a). For viscoelastic heterogeneous media of several discrete linear viscoelastic phases with known stress-strain relations, it was shown that the effective relaxation and creep functions could be obtained by the correspondence principle of the theory of linear viscoelasticity (Hashin, 1965b). In some cases explicit results in terms of general linear viscoelastic matrix properties was given, and thus permitting direct use of experimental information (Hashin, 1966). In a review by Ahmed and Jones (1990) of particulate reinforcement theories for polymer composites, it was concluded that the macroscopic behavior was affected by the size, shape, distribution, and interfacial adhesion of the inclusions. This article makes use of some of these results for elastic composites to explore accessible regions of the stiffness-loss maps of the materials.

ELASTIC AND VISCOELASTIC PROPERTIES OF COMPOSITES

For the simplest case of a two-phase composite, the Voigt and Reuss composites described below represent rigorous upper and lower bounds on the Young's modulus for a given volume fraction of one phase. The Hashin-Shtrikman composites represent upper and lower bounds for isotropic elastic composites. Viscoelastic composites containing spherical or platelet inclusions are also considered. Results obtained via the correspondence principle are plotted as "stiffness-loss maps" in the subsequent section.

Voigt composite

Let phase 1 be stiff; let phase 2 be high loss. The geometry of the Voigt model structure is shown in Figure 2. The composite can contain laminations as shown in Figure 1(a) or it can be made of continuous fibers in Figure 1(b); in either case the strain in each phase is the same. For an elastic material with one of these structures, the Voigt relation is

$E_c = E_1 V_1 + E_2 V_2$,

in which E_c, E_1 and E_2 refer to the Young's modulus of the composite, phase 1 and phase 2, and V_1 and V_2 refer to the volume fraction of phase 1 and phase 2 with $V_1+V_2=1$. The Voigt relation for the stiffness of an elastic composite is obtained by recognizing that for the given geometry, the strain in each phase is the same; the forces in each phase are additive.

By the correspondence principle (Hashin, 1970, Christensen, 1980), the elastic relation can be converted to a steady state harmonic viscoelastic relation by replacing the Young's moduli E by $E^*(i\omega)$ or E^*, in which ω is the angular frequency of the harmonic loading. This procedure gives

$$E_c^* = E_1^* V_1 + E_2^* V_2 \quad (1)$$

with $E^* = E' + i E''$ and loss tangent $\tan\delta = E''/E'$. Taking the ratio of real and imaginary parts, we obtain the loss tangent of the composite $\tan\delta_c = E_c''/E_c'$.

$$\tan\delta_c = \frac{V_1 \tan\delta_1 + V_2 \frac{E_2'}{E_1'} \tan\delta_2}{V_1 + \frac{E_2'}{E_1'} V_2} \tag{2}$$

Reuss composite

The geometry of the Reuss model structure is shown in Figure 1(c); each phase experiences the same stress. For elastic materials, $1/E_c = V_1/E_1 + V_2/E_2$. Again using the correspondence principle, the viscoelastic relation is obtained as

$$\frac{1}{E_c^*} = \frac{V_1}{E_1^*} + \frac{V_2}{E_2^*} \tag{3}$$

Again separating the real and imaginary parts of E_c^*, the loss tangent of the composite $\tan\delta_c$ is obtained:

$$\tan\delta_c = \frac{(\tan\delta_1+\tan\delta_2)[V_1+V_2\frac{E_1'}{E_2'}]-(1-\tan\delta_1\tan\delta_2)[V_1\tan\delta_2+V_2\tan\delta_1\frac{E_1'}{E_2'}]}{(1-\tan\delta_1\tan\delta_2)[V_1+V_2\frac{E_1'}{E_2'}]+(\tan\delta_1+\tan\delta_2)[V_1\tan\delta_2+V_2\tan\delta_1\frac{E_1'}{E_2'}]} \tag{4}$$

Hashin-Shtrikman composite: arbitrary two-phase geometry

Allowing for 'arbitrary' phase geometry, the upper and lower bounds on the elastic moduli as a function of composition have been developed using variational principles. The lower bound for the shear modulus G_L of the composite was given as (Hashin and Shtrikman, 1963)

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

in which K_1, G_1 and V_1, and G_2 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(5) results in the upper bound G_U for the shear modulus.

As for viscoelastic materials, we again apply the correspondence principle. The complex viscoelastic shear moduli of the composite G_L^* and G_U^* are obtained as

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

and

$$G_U^* = G_1^* + \frac{V_2}{\frac{1}{G_2^*-G_1^*} + \frac{6(K_1^*+2G_1^*)V_1}{5(3K_1^*+4G_1^*)G_1^*}} \quad (7)$$

In these cases the loss tangent is more complicated to write explicitly, so it is more expedient to graphically display computed numerical values.

Hashin transversely isotropic fiber reinforced composites

This case is of interest since it allows more than two phases, a situation applicable to the analysis of experimental results in a companion article. For two phases the results are almost identical to the arbitrary phase geometry case considered above. The shear modulus of elastic multi-phase transversely isotropic fiber reinforced composites of arbitrary transverse phase geometry, can be bounded from below and above in terms of phase moduli and phase volume fractions. The lower and upper bounds on the shear modulus $m^{(-)}$ and $m^{(+)}$ were given for elastic composites (Hashin, 1965a) as

$$m^{(-)} = G_1 + \frac{2G_1(K_1+G_1)}{K_1+2G_1}\left\{\left[\sum_{r=2}^{r=n} \frac{(G_r-G_1)V_r}{G_1+K_1G_1/(K_1+2G_1)}\right]^{-1}-1\right\}^{-1} \quad (8)$$

and

$$m^{(+)} = G_n + \frac{2G_n(K_n+G_n)}{K_n+2G_n}\left\{\left[\sum_{r=1}^{r=n-1} \frac{(G_r-G_n)V_r}{G_r+K_nG_n/(K_n+2G_n)}\right]^{-1}-1\right\}^{-1} \quad (9)$$

in which n is the number of the phases, G_1 and K_1 are the shear and bulk moduli of the most compliant phase, G_n and K_n are the shear and bulk moduli of the stiffest phase. r is a free index representing the phase number; phases are numbered in order of increasing stiffness.

On the basis of the correspondence principle, corresponding results for the complex shear modulus (not necessarily bounds) of the composites are again obtained by replacing m(-), m(+), G_1, K_1, G_r and G_n by G_L^*, G_U^*, G_1^*, K_1^*, G_r^* and G_n^* in Equations(8) and (9), respectively. The loss tangent again is complicated to write explicitly, so it is graphically displayed using computed numerical values.

Spherical particulate inclusions

For a small volume fraction $V_1= 1 - V_2$ of spherical elastic inclusions in a continuous phase of another elastic material, the shear modulus of the composite G_c was given as(Christensen, 1979)

$$\frac{G_c}{G_1} = 1 - \frac{15(1-\nu_1)\left(1 - \frac{G_2}{G_1}\right)V_2}{7 - 5\nu_1 + 2(4 - 5\nu_1)\frac{G_2}{G_1}} \quad (10)$$

in which ν_1 is the Poisson's ratio of phase 1, and phase 1 and phase 2 represent the matrix material and the inclusion material respectively.

Using the correspondence principle again and assuming there is no relaxation in Poisson's ratio, Equation(10) becomes

$$G_c^* = G_1^* - \frac{15(1-\nu_1)\ (G_1^* - G_2^*)V_2}{7 - 5\nu_1 + 2(4 - 5\nu_1)\ \frac{G_2^*}{G_1^*}} \quad (11)$$

for the complex shear modulus of the composite material. The loss tangent again is complicated to write explicitly, so it is graphically displayed using computed numerical values.

Platelet inclusions

For a dilute suspension of platelet elastic inclusions of phase 2 in a matrix of phase 1, the shear modulus of the composite G_c was given as(Christensen, 1979)

$$G_c = G_1 + \frac{V_2\ (G_2-G_1)}{15}\left[\frac{9K_2+4(G_1+2G_2)}{K_2 + \frac{4}{3}G_2} + 6\frac{G_1}{G_2}\right] \quad (12)$$

Again, using the correspondence principle, Equation(12) becomes

$$G_c^* = G_1^* + \frac{V_2\ (G_2^*-G_1^*)}{15}\left[\frac{9K_2^*+4(G_1^*+2G_2^*)}{K_2^* + \frac{4}{3}G_2^*} + 6\frac{G_1^*}{G_2^*}\right] \quad (13)$$

for the complex shear modulus of the composite materials.

As for procedure, we remark that although Equations (5) to (13) were developed for the shear modulus of the composite, the shear moduli G^* were replaced by the Young's moduli E^* in the figures for comparison with Fig. 1. The Voigt and Reuss relations given by Equations (1) and (3) apply to G^* as well as to E^*. The actual relationship between E^* and G^* and the properties of the constituents of a composite is simple only for certain phase geometries. For example, for some common phase geometries, a Poisson's ratio of 0.3 for each phase gives a Poisson's ratio close to or equal to 0.3 for the composite. However for some phase geometries, a constituent Poisson's ratio of 0.3 can give rise to a negative Poisson's ratio in cellular solids with one phase void (Lakes, 1987) or in unusual laminates (Milton, in press). The calculations are on the basis that $V_1+V_2=1$ except that $V_1+V_2=0.8$ for the multi-phase Hashin elastic bound, for which 20% void by volume fraction is assumed to be contained as a third phase in the composite.

RESULTS AND DISCUSSION

Results are plotted as stiffness-loss maps (plots of $|E^*|$ vs tan δ) as shown in Figures 2-4 below.

Figure 2 shows predicted properties of composites containing phases which differ greatly in properties. Steel is considered as phase 1, with $|E_1^*|=200$ GPa, tan $\delta_1 = 0.001$ and and a viscoelastic elastomer as phase 2, with $|E_2^*|=0.020$ GPa, tan $\delta_2 = 1.0$. The graph was enlarged in the vicinity of 100% phase 1 and shown in Figure 3 for clarity. A small volume fraction of phase 2 results in a large increase in loss with little reduction in stiffness so that the Reuss structure permits higher losses than the Voigt structure. However, in the Reuss structure each phase carries the full stress, so that a composite of this type will not be strong if, as is usual, the soft phase 2 is weak.

As for 'bounds' on the properties, the curves for the Voigt and Reuss composites enclose a region in the stiffness-loss map, as do the curves for the upper and lower Hashin-Shtrikman composites. It is tempting to think of these curves as 'bounds' on the viscoelastic behavior, however such a surmise has not been proven. They represent extremes of composites which can be fabricated, however we do not yet know if they represent true bounds. Roscoe (1969) has mathematically established bounds for the real and imaginary parts E' and E'' of the complex modulus of composites and has shown them to be equivalent to the Voigt and Reuss relations. Therefore the stiffness, expressed as $|E^*|$ of the composite is bounded from above by the Voigt limit and cannot exceed the stiffness of the stiff phase. This is not quite the same as establishing bounds for a stiffness-loss map since it is not obvious whether a maximum in $\tan \delta = E''/E'$ could be obtained simultaneously with a maximum in E' . In particular, we can construct $\tan \delta_c = E''_{voigt}/E'_{reuss} > E''_{reuss}/E'_{reuss}$ and be within the bounds of Roscoe. We do not yet know if such a composite is physically realizable.

In the stiffness-loss map, the lower and upper two-phase Hashin composites behave similarly to the Voigt and Reuss composites, respectively. This is in contrast to the usual plots of elastic stiffness vs volume fraction, in which the Hashin bounds can differ greatly from the Voigt/Reuss ones. As for the physical attainment of the Voigt and Reuss composites, simple laminates can be made as in Fig. 1, but these are anisotropic. Isotropic composites which attain the Voigt or Reuss moduli are not considered to be attainable. Isotropic polycrystals attaining the Voigt or Reuss bounds for the bulk modulus are also possible (Avellaneda and Milton, 1989) at the expense of some added structural complexity.

For the three-phase Hashin structure with 20% void content in the composite, the lower curve reduces to zero and is not shown in the graph; the upper bound lies close to the Voigt curve with 20% to 40% lower stiffness as shown in Fig. 2.

The composite containing soft spherical inclusions is also found to behave similarly to the Voigt composite in that a small volume fraction of soft, viscoelastic material has a minimal effect on the loss tangent, though it does reduce the stiffness. As for the composite containing soft platelet inclusions, it is found that the results are similar to those of the Reuss structure. A small volume fraction of platelet inclusions as phase 2 results in a very large increase in loss tangent without any significant reduction in the stiffness. However, soft platelets resemble penny-shaped cracks in the matrix, so that such a composite would be weaker than the matrix, particularly if the matrix were brittle.

Figure 4 shows predicted properties of composites containing phases which do not differ so much in properties as steel and viscoelastic elastomers. Copper as phase 1, with $|E_1^*|$=117 GPa, $\tan\delta_1$= 0.002 and indium as phase 2, with $|E_2^*|$=10.8 GPa, $\tan\delta_2$=0.025 (at 1 kHz) were used for this investigation. Observe that the *shape* of this stiffness-loss map differs from the case of the polymer-metal composite. The implication of this difference in shape is as follows. If the constituents differ by orders of magnitude in stiffness and loss, then the Reuss and platelet composites are orders of magnitude superior to the Voigt and spherical inclusion composites in achieving high stiffness and high loss. If the constituents do not differ so much in their properties, their composites of various structures do not differ as much either. Composites containing a stiff, low loss material (such as a metal) and a small amount of compliant, high loss material can exhibit a stiffness close to that of the metal, as well as high loss superior to that of a metal-metal composite.

An interesting aspect of the Reuss and platelet composites which give the highest loss (for given stiffness) is that they exhibit highly nonuniform strain fields. The strain in the soft, lossy phase is much larger than the strain in the stiff phase. This is in contrast to the Voigt composite in which the strain in each phase is the same. The re-entrant foams (Chen and Lakes, 1989, in press) with a negative Poisson's ratio also exhibit non-affine deformation of a more complex nature in that the foam cells unfold as the foam is deformed.

CONCLUSIONS

1. In a stiffness-loss map, the upper and lower two-phase Hashin composites behave similarly to the Voigt and Reuss composites, respectively.
2. Reuss laminates and platelet filled materials based on a stiff, low loss phase and a compliant high loss phase were found to exhibit high stiffness combined with high loss tangent. However, in the Reuss structure each phase carries the full stress, so that a composite of this type will not be strong if, as is usual, the compliant phase is weak.
3. A composite containing soft lossy spherical inclusions in a stiff matrix behaves similarly to the Voigt composite: low loss and a reduction in stiffness.
4. Composites containing a metal and a small amount of compliant, high loss polymer can in principle exhibit a stiffness close to that of the metal, as well as high loss.

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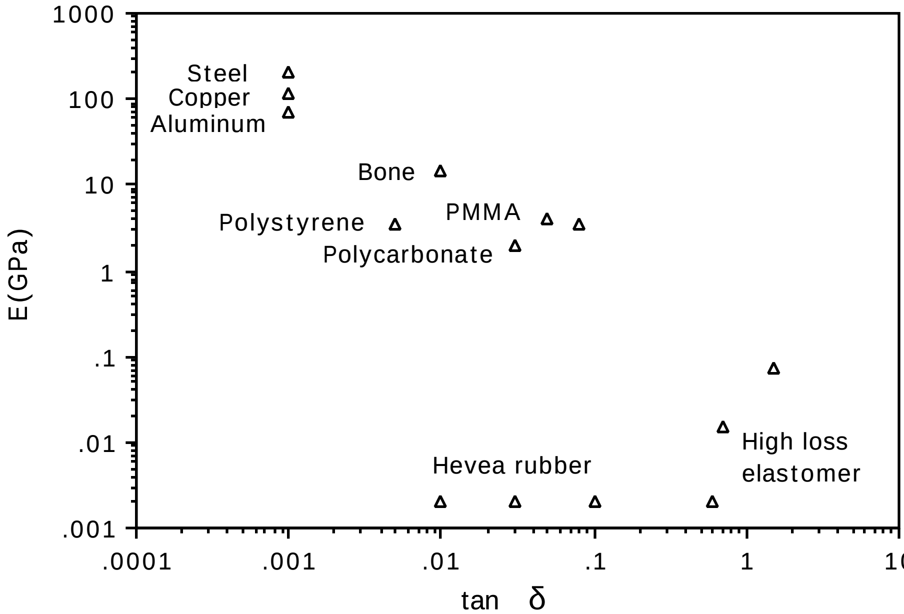
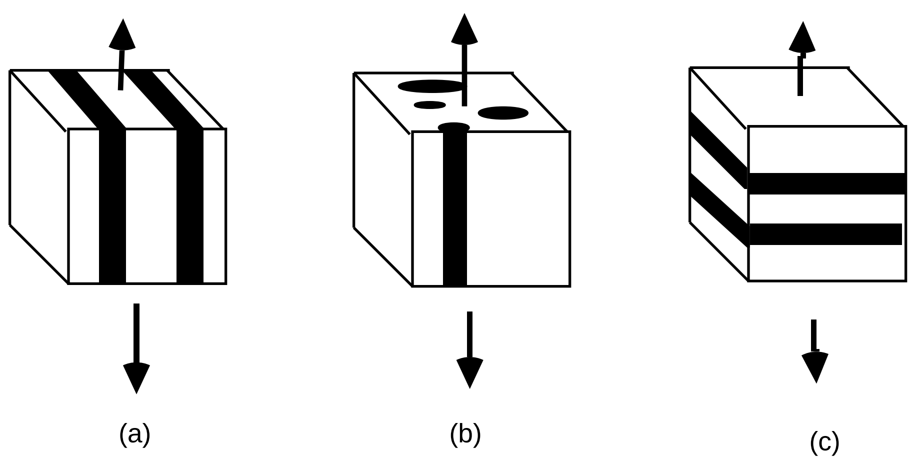
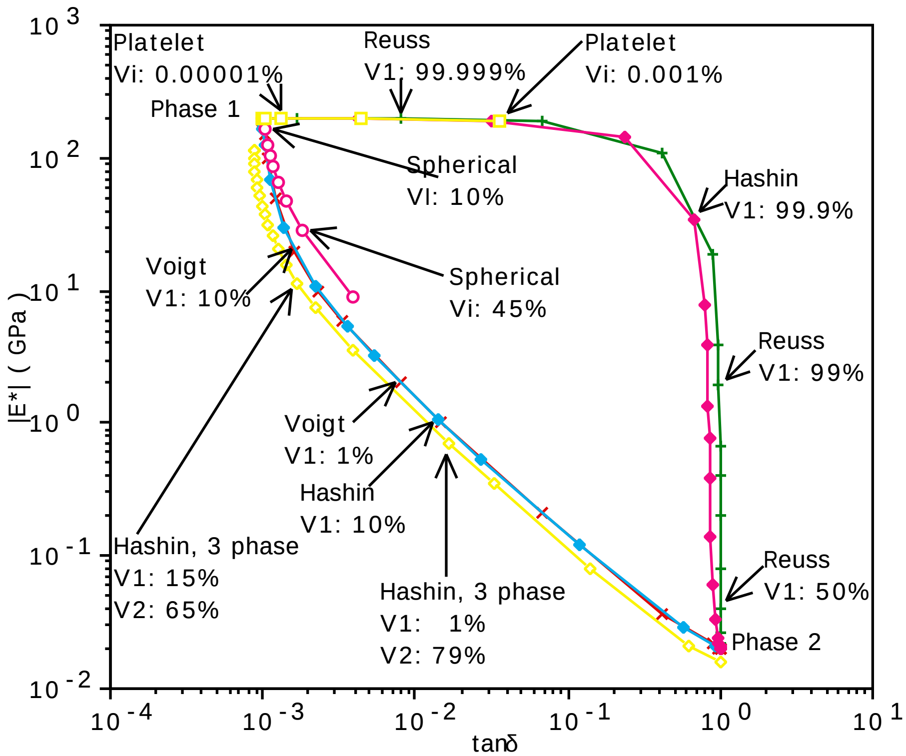
Figures



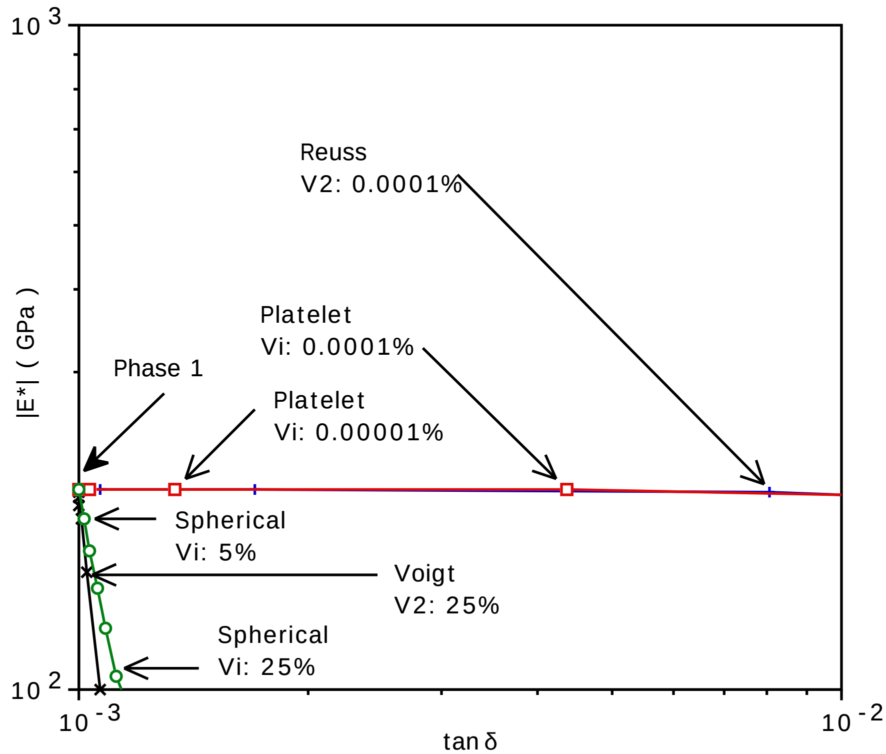
1. Stiffness vs loss tangent for some representative materials at or near room temperature.
   Steel, 1 Hz, after Nowick and Berry (1972)
   Copper, 600 Hz, after Nowick and Berry (1972)
   Polymethyl methacrylate, (PMMA), 10 Hz and 1 kHz, after Ferry (1979)
   Bone, 1-100 Hz, after Lakes, Katz, Sternstein (1979)
   Hevea rubber, 10 Hz-2 kHz, after Ferry (1979)
   Polystyrene, 100 Hz, 1kHz, after Ferry (1979)
   Polycarbonate, 100 Hz, after Nielsen (1962)
   Viscoelastic elastomer, 100, 1,000 Hz, after Shipkowitz, et. al. (1988)



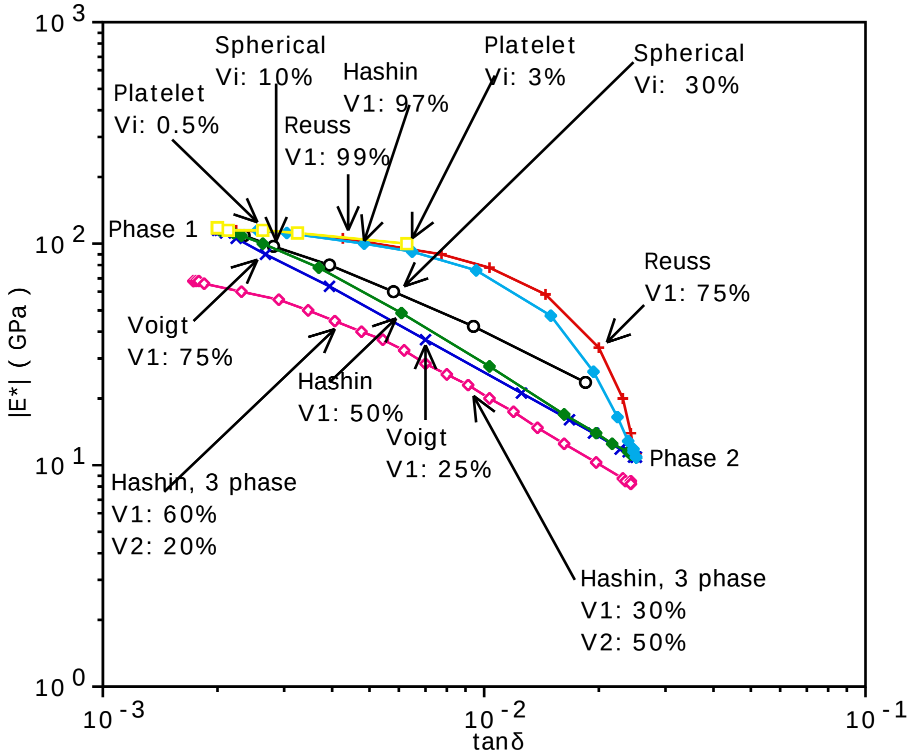
2. (a) Laminated Voigt structure.
   (b) Fibrous Voigt structure.
   (c) Reuss structure.



3. Stiffness-loss map for composites of steel as phase 1 and viscoelastic elastomer as phase 2. x: Voigt curve, +: Reuss curve, ♦ : two-phase Hashin curve, ◊: upper curve of three-phase Hashin composite with 20% voids as one phase. o: composite with phase 2 as dilute spherical inclusions. _ : composite with phase 2 as dilute platelet inclusions.



4. Stiffness-loss map for composites of steel as phase 1 and viscoelastic elastomer as phase 2; expanded plot of upper left portion of Fig. 3. x: Voigt curve, +: Reuss curve, ◊: upper curve of three-phase Hashin composite with 20% voids as one phase. o: composite with phase 2 as dilute spherical inclusions. _ : composite with phase 2 as dilute platelet inclusions.



5. Stiffness-loss map for composites of copper as phase 1 and indium as phase 2. x: Voigt bound, +: Reuss curve, ♦: two-phase Hashin curve, ◊: upper curve of three-phase Hashin composite with 20% voids as one phase. o: composite with phase 2 as dilute spherical inclusions. _ : composite with phase 2 as dilute platelet inclusions.