PHYSICS OF VISCOELASTIC COMPOSITES

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Common Averages

- MEAN:

$$Q_{mean} = \langle Q \rangle = \sum f_i Q_i$$

- HARMONIC MEAN:

$$Q_{harm} = \left\langle \frac{1}{Q} \right\rangle^{-1} = \left[\sum \frac{f_i}{Q_i} \right]^{-1}$$

- GEOMETRIC MEAN:

$$log\, Q_{geo} = \langle log\, Q \rangle = \sum f_i\, log\, Q_i \quad \text{OR}$$

$$Q_{geo} = \Pi_i Q_i^{f_i}$$

Note: $\sum_i f_i = 1$ (for our applications f_i is typically the volume fraction of the i-th constituent).

EXAMPLE:
Wood's Formula for Mixed Fluids

Reuss average (volume weighted harmonic mean) of the bulk modulus:

$$\frac{1}{K_R} = \frac{f_1}{K_1} + \frac{(1-f_1)}{K_2}$$

Volume average of the density:

$$\rho^* = f_1\rho_1 + (1 - f_1)\rho_2$$

Acoustic wave speed at low frequencies and long wavelengths:

$$v_{Wood} = \sqrt{K_R/\rho^*}$$

Related Topics

- Realizability
  - Optimality of bounds
  - Applicability of estimates

- Analytic methods (Bergman-Milton)
  - Bounds (using series expansion analysis)
  - Estimates

- Cross-property bounds

Canonical Functions

- **For electrical conductivity:**

$$\Sigma(s) = \left\langle \frac{1}{\sigma(\vec{r})+2s} \right\rangle^{-1} - 2s$$

- **For bulk modulus:**

$$\Lambda(u) = \left\langle \frac{1}{K(\vec{r})+\frac{4}{3}u} \right\rangle^{-1} - \frac{4}{3}u$$

- **For shear modulus:**

$$\Gamma(z) = \left\langle \frac{1}{\mu(\vec{r})+z} \right\rangle^{-1} - z$$

General Inequalities (for Real Quantities)

- **For electrical conductivity:**

$$0 \le \sigma_{min} \le \left\langle \sigma(\vec{r})^{-1} \right\rangle^{-1} \le \sigma_{eff} \le \langle \sigma(\vec{r}) \rangle \le \sigma_{max} \le \infty$$

- **For bulk modulus:**

$$0 \le K_{min} \le \left\langle K(\vec{r})^{-1} \right\rangle^{-1} \le K_{eff} \le \langle K(\vec{r}) \rangle \le K_{max} \le \infty$$

- **For shear modulus:**

$$0 \le \mu_{min} \le \left\langle \mu(\vec{r})^{-1} \right\rangle^{-1} \le \mu_{eff} \le \langle \mu(\vec{r}) \rangle \le \mu_{max} \le \infty$$

Hashin-Shtrikman Bounds (1)

- **For electrical conductivity:**

$$\sigma^{-}_{HS} \leq \sigma_{eff} \leq \sigma^{+}_{HS},$$

where

$$\sigma^{-}_{HS} = \Sigma(\sigma_{min}) \text{ and } \sigma^{+}_{HS} = \Sigma(\sigma_{max})$$

- **For bulk modulus:**

$$K^{-}_{HS} \leq K_{eff} \leq K^{+}_{HS},$$

where

$$K^{-}_{HS} = \Lambda(\mu_{min}) \text{ and } K^{+}_{HS} = \Lambda(\mu_{max})$$

Hashin-Shtrikman-Walpole Bounds (2)

- **For shear modulus:**

$$\mu_{HS}^{-} \leq K_{eff} \leq \mu_{HS}^{+},$$

where

$$\mu_{HS}^{-} = \Gamma(\zeta_{min}) \text{ and } \mu_{HS}^{+} = \Gamma(\zeta_{max})$$

with

$$\zeta_{min} = \frac{\mu_{min}}{6}\frac{9K_{min}+8\mu_{min}}{K_{min}+2\mu_{min}}$$

$$\zeta_{max} = \frac{\mu_{max}}{6}\frac{9K_{max}+8\mu_{max}}{K_{max}+2\mu_{max}}.$$

HS only considered well-ordered case $(K_1 - K_2)(\mu_1 - \mu_2) > 0$.

Walpole considered general case, so these are called HSW bounds.

OUTLINE

- Review of Some Averaging Methods
- Bounds for Real Elastic Constants
- Effective Medium Theories, including the CPA
- Bounds for Complex Viscoelastic Constants
- Conclusions

Self-Consistent Method or CPA

- **For electrical conductivity:**

$$\sigma^* = \Sigma(\sigma^*)$$

- **For elastic constants (K and $G \equiv \mu$):**

$$K^* = \Lambda(G^*)$$
$$G^* = \Gamma(\zeta^*),$$

where

$\zeta^* = (G^*/6)(9K^* + 8G^*)/(K^* + 2G^*)$ depends on both K^* and G^*.

Elastic Wave Scattering Theory (1)

Take a plane compressional wave with displacement $\vec{u}$

$$\vec{u} = \hat{x}(ik)^{-1}\, exp\, i(kx - \omega t),$$

incident along the x-axis on a spherical scatterer (an inhomogeneity in bulk modulus, density, and/or shear modulus). The radial component of the scattered wave is

$$(u_r^s)_i = (ik)^{-1}(ka_i)^3\, exp\, i(kr_i - \omega t)/kr_i$$

$$\times[B_0 - B_1\, cos\,\theta - B_2(3\, cos\, 2\theta + 1)/4],$$

and the transverse component is

$$(u_t^s)_i = (ik)^{-1}(sa_i)^3\, exp\, i(sr_i - \omega t)/sr_i$$

$$\times[B_1\, sin\,\theta + (3s/4k)B_2\, sin\, 2\theta],$$

Elastic Wave Scattering Theory (2)

where r_i is measured from the center of the spherical scatterer i, and a_i is the radius of the sphere, while

$$s = \omega(\rho_m/G_m)^{1/2}$$

and

$$k = \omega[\rho_m/(K_m + 4G_m/3)]^{1/2}$$

are, respectively, the wavenumbers for shear and compressional waves in the host matrix (m).

Coherent Potential Approximation for Elasticity

Low Frequency Scattering Coefficients: Monopole, Dipole, and Quadupole

$$B_0(K_m, K_i, G_m) = \frac{K_m - K_i}{3K_i + 4G_m},$$

$$B_1(\rho_m, \rho_i) = (\rho_m - \rho_i)/3\rho_m,$$

$$B_2(G_m, G_i, K_m) = \frac{20G_m(G_i - G_m)/3}{6G_i(K_m + 2G_m) + G_m(9K_m + 8G_m)}.$$

Host medium uses subscript m, inclusions i.

Inclusion is a spherical scatterer imbedded in the host for the present argument.

Derivation of the CPA Formulas

The composite (scattering) medium is imbedded in an adjustable matrix material $m = *$, such that each individual scatterer sees all the other scatterers as composing this matrix. Then, the composite inclusion, when imbedded in the $*$-matrix, should actually produce no scattering at all at infinity if the single-scattering coefficients satisfy

$$\sum_{i=1}^{n} f_i B_0(K^*, K_i, G^*) = 0,$$

$$\sum_{i=1}^{n} f_i B_2(G^*, G_i, K^*) = 0.$$

CPA Formulas

The final results we obtain using the CPA are

$$\frac{1}{K^{eff}+4G^{eff}/3} = \sum_{i=1}^{n} \frac{f_i}{K_i+4G^{eff}/3} = \left\langle \frac{1}{K(x)+4G^{eff}/3} \right\rangle,$$

and

$$\frac{1}{G^{eff}+\zeta^{eff}} = \sum_{i=1}^{n} \frac{f_i}{G_i+\zeta^{eff}} = \left\langle \frac{1}{G(x)+\zeta^{eff}} \right\rangle,$$

where $\zeta \equiv G(9K+8G)/6(K+2G)$. Note that these equations are coupled and must be solved iteratively. The notation $\langle \cdot \rangle$ is just the volume average.

The Kuster-Toksöz Approximation in Elasticity

Low Frequency Scattering Coefficients

$$B_0(K_m, K_i, G_m) = \frac{K_m - K_i}{3K_i + 4G_m}$$

$$B_2(G_m, G_i, K_m) = \frac{20G_m(G_i - G_m)/3}{6G_i(K_m + 2G_m) + G_m(9K_m + 8G_m)}$$

Host medium uses subscript m, inclusions i.

Inclusion is a spherical scatterer imbedded in the host for the present argument.

Derivation of the Kuster-Toksöz Formulas

The composite (scattering) medium is imbedded in a fixed host material $m = h$, where h is any one of the constituents in the composite. Then, the composite inclusion of type-$*$, when imbedded in the h-matrix, should produce the correct amount of scattering at infinity if the single-scattering coefficients satisfy

$$B_0(K_h, K^*, G_h) = \sum_{i=1}^{n} f_i B_0(K_h, K_i, G_h),$$

$$B_2(G_h, G^*, K_h) = \sum_{i=1}^{n} f_i B_2(G_h, G_i, K_h).$$

Kuster-Toksöz or ATA Formulas

The final results we obtain using the Kuster-Toksoz approach are

$$\frac{1}{K^*+4G_h/3} = \sum_{i=1}^{n} \frac{f_i}{K_i+4G_h/3} = \left\langle \frac{1}{K(x)+4G_h/3} \right\rangle,$$

and

$$\frac{1}{G^*+\zeta_h} = \sum_{i=1}^{n} \frac{f_i}{G_i+\zeta_h} = \left\langle \frac{1}{G(x)+\zeta_h} \right\rangle,$$

where $\zeta \equiv G(9K+8G)/6(K+2G)$. Note that for spherical inclusions these equations are not coupled. If the host material is chosen to be one having either the largest or smallest constants, then Kuster-Toksoz gives the same results as the Hashin-Shtrikman bounds.

Canonical Functions of Elasticity

For bulk modulus,

$$\Lambda(G) \equiv \left\langle \frac{1}{K(x)+4G/3} \right\rangle^{-1} - \frac{4}{3}G.$$

For shear modulus,

$$\Gamma(\zeta) \equiv \left\langle \frac{1}{G(x)+\zeta} \right\rangle^{-1} - \zeta,$$

where

$$\zeta \equiv G(9K + 8G)/6(K + 2G).$$

Monotonicity of the Canonical Functions

Both canonical functions are monotonic functions of their arguments.

For example,

$$\frac{d\Lambda(G)}{dG} = \frac{4}{3}\left\langle \frac{1}{K(x)+4G/3} \right\rangle^{-2} \times$$

$$\left(\left\langle \frac{1}{(K(x)+4G/3)^2} \right\rangle - \left\langle \frac{1}{K(x)+4G/3} \right\rangle^2 \right) \geq 0.$$

Non-negativity follows easily from the Cauchy-Schwartz inequality $\langle a\rangle^2 \leq \langle a^2\rangle$.

Effective Medium Theory Results

CPA results for bulk modulus are

$$K^{eff} = \Lambda(G^{eff}).$$

CPA results for shear modulus are

$$G^{eff} = \Gamma(\zeta^{eff}).$$

Note that the two equations are coupled, and must be iterated to obtain their solutions.

Hashin-Shtrikman Bounds

Hashin-Shtrikman bulk modulus bounds are

$$K_{HS}^{\pm} = \Lambda(G_{\pm}).$$

Hashin-Shtrikman shear modulus bounds are

$$G_{HS}^{\pm} = \Lambda(\zeta_{\pm}),$$

where $K_+ = max_i\, K_i$, $K_- = min_i\, K_i$, $G_+ = max_i\, G_i$, $G_- = min_i\, G_i$, and $\zeta \equiv G(9K + 8G)/6(K + 2G)$.

Complex Bulk Modulus Bounds (1)

Define four points in the complex plane, using the complex version of the canonical functions, i.e., same definition of the function but the bulk moduli appearing in the expression can now be considered complex:

$$K_{1*} = \Lambda(G_1)$$

$$K_{2*} = \Lambda(G_2)$$

$$K_{h*} = \Lambda(0)$$

$$K_{a*} = \Lambda(\infty).$$

And then recall that K_1 and K_2 are the values of the complex bulk moduli of the constituents.

Complex Bulk Modulus Bounds (2)

Then, we define four arcs in the complex K-plane, each arc passing through the first two points, and also through one of the remaining four points according to:

$$\mathrm{Arc}(K_{1*}, K_{2*}, K_{h*}), \quad \mathrm{Arc}(K_{1*}, K_{2*}, K_{a*}),$$
$$\mathrm{Arc}(K_{1*}, K_{2*}, K_{1}), \quad \mathrm{Arc}(K_{1*}, K_{2*}, K_{2}),$$

where

$$\mathrm{Arc}(\alpha_1, \alpha_2, \alpha_3) \equiv \gamma_1\alpha_1 + \gamma_2\alpha_2 - \frac{\gamma_1\gamma_2(\alpha_1-\alpha_2)^2}{\gamma_2\alpha_1+\gamma_1\alpha_2-\alpha_3},$$

and $\gamma_1 = 1 - \gamma_2$ is real and varies along [0,1].

This is a linear fractional, or bilinear, transformation.

Some Theoretical Development (1)

Defining complex strain ϵ, complex stress σ, and complex stiffness C (which varies in space because of the presence of two distinct constituents), we have the viscoelastic constitutive relationship:

$$\sigma = C\varepsilon.$$

Think of this as a 6×6 system of equations: σ and ε are each 1×6 complex vectors, and C is a 6×6 complex matrix (using the Voigt convention for transforming elastic or viscoelastic tensors to matrices).

Some Theoretical Development (2)

If real parts are distinguished by single primes and imaginary parts by double primes, then we can define real, composite vectors

$$j = \begin{pmatrix} \varepsilon'' \\ \sigma'' \end{pmatrix} \quad \text{and} \quad e = \begin{pmatrix} -\sigma' \\ \varepsilon' \end{pmatrix},$$

and a corresponding 12×12 system

$$j = De,$$

where now

$$D = \begin{pmatrix} (C'')^{-1} & (C'')^{-1}C' \\ C'(C'')^{-1} & C'' + C'(C'')^{-1}C' \end{pmatrix}$$

is a 12×12 real matrix.

Some Theoretical Development (3)

So, there is still a lot of work to do, but now we are dealing just with real quantities. We have to analyze the behavior of the average D matrix for our binary composite, but this is relatively straightforward now within the context of the random media theory. We can just generalize the Hashin-Shtrikman variational principle for this more general problem. This results in bounds on both complex bulk modulus and complex shear modulus.

We are going to skip over these details here, so I can show you some related developments concerning the canonical functions and the so-called Y-tensor.

Canonical Functions and the Y-Tensor (1)

Let the actual effective constants be K^* and G^*.

Then, with the definitions given before of Λ and Γ,

we can define arguments Y_K^* and Y_G^* such that

$$K^* = \Lambda(3Y_K^*/4) \quad \text{and} \quad G^* = \Gamma(Y_G^*).$$

Now, the canonical functions are one-to-one and can be inverted

for their arguments in terms of the other quantities:

$$Y_K^* = \frac{K_1 K_2 (K^* <1/K> - 1)}{<K> - K^*},$$

$$Y_G^* = \frac{G_1 G_2 (G^* <1/G> - 1)}{<G> - G^*}.$$

This fact is very useful because, if we can bound the Y's,

then we can obtain rather tight bounds on K^* and G^*.

Canonical Functions and the Y-Tensor (2)

In the present context, we can define the general Y^*-tensor according to:

$$Y^*(D^*, D_1, D_2) = \begin{pmatrix} (y'')^{-1} & -(y'')^{-1}y' \\ -y'(y'')^{-1} & y'' + y'(y'')^{-1}y' \end{pmatrix},$$

where the complex 6×6 matrix y is defined by

$$y(C^*, C_1, C_2) =$$
$$-f_2C_1 - f_1C_2 + f_1f_2(C_1 - C_2)(f_1C_1 + f_2C_2 - C^*)^{-1}(C_1 - C_2).$$

This matrix can be broken down into its bulk and shear parts.

Canonical Functions and the Y-Tensor (3)

When this has been done, we ultimately $(\ldots)$ are able to show that:

$$\begin{pmatrix} \frac{1}{2y''_G} & -\frac{y'_G}{y''_G} \\ -\frac{y'_G}{y''_G} & 2\left[y''_G + \frac{(y'_G)^2}{y''_G}\right] \end{pmatrix} - Z \geq 0.$$

Here Z is a 2×2 real comparison matrix. This statement says that the spectrum of the 2×2 matrix on the left must be nonnegative. So its eigenvalues must be nonnegative. After some more math, we can show that this implies the point $y'_G + iy''_G$ must lie inside certain circles in the complex Y_G-plane. The matrix Z contains some parameters that we are free to vary, and the final bounds lie inside the convex hull generated by considering all these bounding circles.

Complex Hashin-Shtrikman-Walpole Points

Just as in the case of real coefficients, the complex shear modulus depends on a combination of the bulk and shear moduli of the constituents. But, unlike the real case, there are four of these points and they are need not fall in any simple order on the complex plane. They are:

$$Y_{11} = y(K_1, G_1) = \tfrac{G_1}{6}\tfrac{9K_1+8G_1}{K_1+2G_1},$$
$$Y_{22} = y(K_2, G_2) = \tfrac{G_2}{6}\tfrac{9K_2+8G_2}{K_2+2G_2},$$
$$Y_{12} = y(K_1, G_2) = \tfrac{G_2}{6}\tfrac{9K_1+8G_2}{K_1+2G_2},$$
$$Y_{21} = y(K_2, G_1) = \tfrac{G_1}{6}\tfrac{9K_2+8G_1}{K_2+2G_1}.$$

We call these the Hashin-Shtrikman-Walpole points, by analogy to the case of coefficients. They turn out to be important points in many cases, as we will see in the examples.

CONCLUSIONS

• Effective medium theories and rigorous bounding methods are rather difficult for viscoelastic constants, but substantial progress has been made in recent years.

• The canonical functions and the related Y-transform methods seem to play a central role in most of the effective medium theory approaches and also in the bounding methods.

• The CPA and differential effective medium theory methods are known to be realizable, i.e., have definite microstructures associated with them, and so are expected always to lie inside rigorous bounds. This has been confirmed in all the examples.