Interfacial tension and a three-phase generalized self-consistent theory of non-dilute soft composite solids

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In the dilute limit Eshelby’s inclusion theory captures the behavior of a wide range of systems and properties. However, because Eshelby’s approach neglects interfacial stress, it breaks down in soft materials as the inclusion size approaches the elastocapillarity length \( L \equiv \gamma/E \). Here, we use a three-phase generalized self-consistent method to calculate the elastic moduli of composites comprised of an isotropic, linear-elastic compliant solid hosting a spatially random monodisperse distribution of spherical liquid droplets. As opposed to similar approaches, we explicitly capture the liquid–solid interfacial stress when it is treated as an isotropic, strain-independent surface tension. Within this framework, the composite stiffness depends solely on the ratio of the elastocapillarity length \( L \) to the inclusion radius \( R \). Independent of inclusion volume fraction, we find that the composite is stiffened by the inclusions whenever \( R < 3L/2 \). Over the same range of parameters, we compare our results with alternative approaches (dilute and Mori–Tanaka theories that include surface tension). Our framework can be easily extended to calculate the composite properties of more general soft materials where surface tension plays a role.

While the effective bulk modulus is determined analytically, there is no exact solution for the effective shear modulus, although considerable information is available on its variational bounds.

In addition to the bounds mentioned above, a broad class of self-consistent (SC) methods have been developed that yield analytical predictions for both bulk and shear moduli. For example, Kroner introduced a self-consistent approximation wherein the inclusion itself is directly embedded in an unknown homogeneous effective medium. Budiansky and Hill use this approach in their model for elastic moduli of composites, although they noted physical inconsistencies at high inclusion volume fractions \( \phi \).

A three-phase generalized self-consistent (GSC) model was introduced by Kerner and van der Poel, Christensen and Lo took this approach by replacing the set of all actual inclusions by a single ideal inclusion, in the “composite spheres” framework described above. Whereas the SC method is generally simpler than GSC models, the proper boundary conditions of the latter remove the unphysical behavior of the former as \( \phi \) becomes large. Both approaches typically assume continuity of displacement (or no slip) across interfaces [e.g. ref. 13], although the bulk modulus has been shown to be unaffected by finite slip.

The surface stress at solid/liquid interfaces can have a substantial range of size-dependent effects in soft materials. For example, recent work has shown that surface stress significantly influences pearling and creasing instabilities, wetting, adhesion, and the relaxation of soft solids towards their equilibrium shapes [e.g., ref. 29]. Hence, here we aim to reformulate the micromechanics

I. Introduction

Composite materials are of interest because they rarely have the bulk properties of their constituents alone. Thus, understanding their properties provides a challenge and a test bed for both controlling material behavior and understanding how and why natural materials have evolved [e.g., ref. 1 and 2 and references therein]. Because in both engineering and natural settings there is always a compromise between the ability to tailor every detail and achieving an optimal effective behavior, such as stiffness, theoretical approaches that span the widest range of key control parameters are desirable.

Among the most successful idealized geometric models for two-phase matrix-inclusion composites is Hashin’s composite-spheres model, where the actual composite is replaced by a set of “composite spheres” with a suitable size-distribution, and arranged in a volume-filling configuration. Each composite sphere consists of a homogeneous sphere representing the inclusion phase, surrounded by a concentric spherical shell of matrix material. The ratio between internal and external radii of each shell is determined in terms of the volume fraction occupied by the inclusion phase within the actual composite.
of soft composites in the non-dilute limit to include the effects of surface stress. In so doing, we systematically examine how the interplay between the inclusion volume fraction \( \phi \) and the inclusion size \( R \) influences the mechanical properties. We note that, for both the dilute and non-dilute cases, the inclusion/matrix surface stress has been treated in previous work, the most notable of which however assumes linear (in-strain) surface stress, and/or uses incorrect boundary conditions, as described previously. 35

Style et al. studied a dilute monodisperse random spatial distribution of liquid droplets of radius \( R \) embedded in a homogeneous isotropic elastic solid matrix. They included the effect of surface tension \( \gamma \) at the inclusion/matrix interface and showed the importance of \( R \) relative to the elastocapillarity length \( \lambda_{EC} \), where \( \lambda_{EC} \) is the Young’s modulus of the matrix. Here, we use a three-phase GSC model to examine the stiffness in such a composite system for finite inclusion volume fractions. We compare the results with the dilute theory and an extension of the Mori–Tanaka theory to non-dilute soft composite solids, both of which include surface tension.

II. The model

Consider a composite system of many identical incompressible droplets embedded in an isotropic homogeneous elastic solid with shear modulus \( \mu_2 \) and Poisson ratio \( \nu_2 \) as shown in Fig. 1. We ask how surface tension at the droplet/matrix interface affects Christensen and Lo’s solution for the effective modulus of the composite for non-dilute droplet volume fractions \( \phi \). Our approach is based on the three-phase self-consistent model of Kerner. 9

As noted above, the GSC approach treats the multi-droplet system as a single composite sphere embedded in an infinite medium of unknown effective elastic moduli \( \mu_s, \nu_s \). The composite sphere consists of a liquid droplet of radius \( R \), surrounded by a concentric spherical shell of matrix material of radius \( R/\phi^{1/3} \), thereby preserving the liquid volume fraction \( \phi \) of the original multi-droplet system. The overall approach follows that of previous work [e.g., ref. 35 and 38], up to a point. To make this paper reasonably self-contained we summarize the key intermediate results, and provide more detail where we believe clarity is required.

Placing the origin of an \((r, \theta, \phi)\) spherical coordinate system at the center of the composite sphere, we choose the following far-field \((r \to \infty)\) displacements:

\[
\begin{align*}
\mathbf{u}_r^0 & = 2e^0_A r \mathbf{P}_2(\cos \theta), \\
\mathbf{u}_\theta^0 & = e^0_A r \frac{d \mathbf{P}_2(\cos \theta)}{d \theta}, \\
\mathbf{u}_\phi^0 & = 0,
\end{align*}
\]

where \( \mathbf{P}_2 \) is the Legendre polynomial of order 2. Note, the azimuthal angle \( \phi \) in eqn (1) is not to be confused with the volume fraction used throughout the remainder of the paper. The corresponding, purely deviatoric far-field strains are

\[
\mathbf{\varepsilon}_r^0 = \mathbf{\varepsilon}_\theta^0 = -\mathbf{\varepsilon}_\phi^0, \quad \mathbf{\varepsilon}_\phi^0 = 2\mathbf{\varepsilon}_A.
\]

For the strained system, the symmetry about the \( z \)-axis allows the use of the following ansatz [e.g., ref. 33, 39] for the displacements \( \mathbf{u}_i^0 \) and \( \mathbf{u}_i^1 \) in the radial and polar directions,

\[
\mathbf{u}_r^1(\rho, \theta) = \left( \mathbf{F}_1 + \frac{\mathbf{G}_1}{\rho^2} \right) r + \mathbf{P}_2(\cos \theta)
\]

\[
\times \left[ 12\nu_1 A_i \rho^2 + 2B_i + 2 \left( \frac{5 - 4\nu_1}{\rho^3} - 3 \frac{\mathbf{D}_i}{\rho^2} \right) r \right],
\]

\[
\mathbf{u}_\theta^1(\rho, \theta) = \frac{d \mathbf{P}_2(\cos \theta)}{d \theta}
\]

\[
\times \left[ (7 - 4\nu_1) A_i \rho^2 + B_i + 2 \left( \frac{1 - 2\nu_1}{\rho^3} - \frac{\mathbf{D}_i}{\rho^2} \right) r \right],
\]

where \( \rho \equiv r/R \), the index “\( i \)” refers to either the matrix \((i = 2)\) or the composite effective medium \((i = 3)\) phase, and \( \mathbf{A}_i, \mathbf{G}_i \) through \( \mathbf{G}_i \) will be determined from the boundary conditions.

The corresponding stress components in regions \( i = 2, 3 \) are

\[
\sigma_{rr}^{(i)}(\rho, \theta) = 2\mu_i \left\{ -\frac{\mathbf{G}_1}{\rho^2} \left( \mathbf{F}_1(1 + \nu_1) \right) \\
+ \left[ -6\nu_1 A_i \rho^2 + 2B_i - \frac{4(5 - \nu_1)}{\rho^3} C_i + \frac{12D_i}{\rho^2} \right] \mathbf{P}_2(\cos \theta) \right\},
\]

and

\[
\sigma_{\theta\theta}^{(i)}(\rho, \theta) = 2\mu_i \frac{d \mathbf{P}_2(\cos \theta)}{d \theta}
\]

\[
\times \left[ (7 + 2\nu_1) A_i \rho^2 + B_i + 2 \left( 1 + \nu_1 \right) C_i + 4\mathbf{D}_i \right].
\]

The relation between the pressure \( p \) and the components of the hydrostatic stress tensor in the liquid region \((i = 3)\) is:

\[
\sigma_{rr}^{(3)} = \sigma_{\theta\theta}^{(3)} = -p, \quad \sigma_{\phi\phi}^{(3)} = 0.
\]

Now, combining the stress-displacement relationship with eqn (1) gives the far-field stresses:

\[
\sigma_{rr}^0 = 4\mu_s \mathbf{P}_2(\cos \theta), \quad \sigma_{\theta\theta}^0 = 2\mu_s \frac{d \mathbf{P}_2(\cos \theta)}{d \theta}.
\]
Three constants are determined by the far-field stress/strain, viz., $A_3 = 0$, $B_3 = \phi_3^0$, and $F_3 = 0$. There are ten equations for the remaining ten unknowns; $p, A_3, B_2, C_2, D_2, F_2, G_2, C_3, D_3, G_3$. Six equations arise from continuity of displacement and stress at the composite sphere surface ($\rho = x \equiv 1/\phi^{1/3}$), one from the incompressibility of the droplet and three from the stress boundary conditions associated with the generalized Young–Laplace condition. Taking these in turn, we have
\[
\begin{align*}
  u_{\theta}^{(2)}(x,0) &= u_{\theta}^{(3)}(x,0), \quad u_{\phi}^{(2)}(x,0) = u_{\phi}^{(3)}(x,0) & \text{and} \\
  \sigma_{\theta}^{(2)}(x,0) &= \sigma_{\theta}^{(3)}(x,0), \quad \sigma_{\phi}^{(2)}(x,0) = \sigma_{\phi}^{(3)}(x,0),
\end{align*}
\]
where the continuity of displacement for $u_\theta, (u_\phi)$ provides two (one) equations and that for stress $\sigma_{\theta}, (\sigma_{\phi})$ provides two (one) equations; droplet incompressibility requires that
\[
\int_{\sin^{-1}} u_{\phi}^{(3)}(1, \theta) R^2 \sin \theta \, d\theta = R^3 (F_2 + G_2) = 0,
\]
where $S^{\text{int}}$ denotes the droplet/matrix interface. The final three equations arise from the stress boundary conditions at the surface of the droplet, treated using the generalized Young–Laplace condition written as
\[
\sigma \cdot n = -p n + \gamma \kappa n,
\]
where $n$ is the normal to the droplet surface and $\kappa$ is its curvature, while the surface stress is taken as an isotropic and strain-independent surface tension, $\gamma$. Using leading-order expressions for $n$ and $\kappa$, eqn (11) becomes
\[
\begin{align*}
  \sigma_{\theta} + \sigma_{\phi} &\left( \frac{\partial u_{\theta}}{\partial \theta} \right) R^{-1} \left( \frac{2 R}{\theta} + \frac{\partial^2 u_{\theta}}{\partial \theta^2} \right) - \left( \frac{u_{\theta} - \partial u_{\theta}}{\partial \theta} \right) R^{-1} = \frac{p + 0}{0 - p} \frac{u_{\theta} - \partial u_{\theta}}{\partial \theta} R^{-1} \\
  \sigma_{\phi} + \sigma_{\phi} &\left( \frac{\partial u_{\phi}}{\partial \phi} \right) R^{-1} \left( \frac{2 R}{\phi} + \frac{\partial^2 u_{\phi}}{\partial \phi^2} \right) - \left( \frac{u_{\phi} - \partial u_{\phi}}{\partial \phi} \right) R^{-1} = \frac{p + 0}{0 - p} \frac{u_{\phi} - \partial u_{\phi}}{\partial \phi} R^{-1} \quad (12)
\end{align*}
\]
where $\gamma$ is the outer sphere as a function of $\phi, \phi_3, \gamma/(\gamma R_3)$ as follows
\[
W = \frac{1}{(3/4)} \left[ \frac{\partial^2 u_{\theta}}{\partial \theta^2} + \frac{\partial^2 u_{\phi}}{\partial \phi^2} \right] + \gamma/(\gamma R_3) = \frac{4\pi \mu_3 R_3^3 \phi_3 (\phi_3 - 1)}{\phi_3^{3/3}}
\]
Therefore, $C_3 = 0$, and plugging this into the solution of the system of eqn (9)–(12) yields a quadratic condition for the relative effective shear modulus $\mu_{rel} = \mu_3/\mu_1$ as a function of $\phi_3$, $\nu_2$, and $\gamma/(\gamma R_3)$ as follows
\[
2\mu_3 (a_0 + a_1 \mu_{rel} + a_2 \mu_{rel}^2) + \gamma(b_0 + b_1 \mu_{rel} + b_2 \mu_{rel}^2) = 0,
\]
where the coefficients are in Appendix A.

For the remainder of the paper we will focus on the special case of an incompressible matrix, for which $E_{rel} = (E_1/E_2) = (\mu_3/\mu_2)$ and $\nu_2 = 1/2$. Considering the elastocapillary length $L \equiv \gamma/E_2$ based on the matrix phase of the composite sphere, we define the dimensionless parameter $\gamma' \equiv L/R = \gamma/(E_2 \gamma)$. Fig. 2 shows the behavior of $E_{rel}$ as a function of $\phi$, and in Fig. 3 $E_{rel}$ is plotted against $R'[(3/4\pi)\gamma^3]$, where $V$ is the outer sphere volume in the GSC framework (i.e., $E_{rel}$ is plotted against $\phi^{1/3}$). Clearly, Fig. 2 shows a monotonic response over a large range of $\phi$, exhibiting softening (stiffening) behavior for
\( g_0^{2/3} \) (\( g_0^{4/3} \)), which spans the experimental range seen by Style et al.\(^{36}\) Moreover, the dilute theory\(^{35}\) is quantitatively captured in the limit \( f \rightarrow 0 \) of the present theory. Furthermore, we find exact “mechanical cloaking”, in which \( E_{\text{rel}} \) is constant at \( g_0^{2/3} \) for all liquid volume fractions. Exactly the same cloaking condition is found in the dilute theory\(^{35}\) and from a complimentary Mori–Tanaka approach,\(^{37}\) which agrees with ref. 41 in the limit of an incompressible matrix, as described in Fig. 4 below.

In the stiffening regime, as droplets become small and \( \gamma' \) becomes large, the quadratic condition (15) for \( \mu_{\text{rel}} \) takes the form \( b_0 + b_1 \mu_{\text{rel}} + b_2 \mu_{\text{rel}}^2 = 0 \). At a given \( \phi \) the solution of this equation, \( \mu_{\text{rel}} = \mu_{\text{rel}, R = L}[\theta, \nu, \phi] \), gives the upper limit of rigidity among all \( \gamma' \)-curves, showing a stiffening behavior proportional to \( \frac{1}{1 - \phi} \) in the limit \( \phi \rightarrow 0 \) (see the \( \gamma' = \infty \) line in Fig. 2).

Now we examine the deformation of the inclusion phase by making use of eqn (B1) and (B2) in Appendix B to evaluate the effective droplet strain \( e_d \) (\( l/r \)) of the droplet.\(^{51}\) In terms of \( x = f_{\text{rel}}^{-1/3}, \gamma' \), and the solution of eqn (15), \( \mu_{\text{rel}} = \mu_{\text{rel}}[\theta, \nu, \phi] \), the radial and polar displacements of the droplet interface are

\[
\frac{u_r(1, \theta)}{R} = 100 \mu_{\text{rel}} x^3 e_0 \frac{f_1}{f_2 + \gamma' f_3} P_2(\cos \theta)
\]

(16)

and

\[
\frac{u_{\theta}(1, \theta)}{R} = 25 \mu_{\text{rel}} x^3 e_0 \frac{f_4}{f_2 + \gamma' f_5} dP_2(\cos \theta).
\]

(17)

where the coefficients \( f_1 \)–\( f_5 \) are in Appendix B. When \( R \ll L \) and \( \gamma' \gg 1 \), and the radial displacement becomes extremely small, then the inclusions remain spherical. In the opposite limit,
$R \gg L$ and $\gamma' \ll 1$, the inclusion shape is again scale-invariant, in agreement with the theory for the case of pure bulk elasticity. The corresponding effective droplet strain is $e_{\text{eff}} = 200\mu_{\text{rel}}x^2\phi_f/(f_2 + \gamma'f_2)$. In the dilute limit ($\phi \to 0$) of the incompressible case, the droplet's effective strain and shape reduce to $e_{\text{eff}}|_{\phi \to 0} = 40\phi/(6 + 15\gamma')$,

$$u_t(1, \theta) = \frac{5}{3} \phi \left(1 + 2 \cos(2\theta) + 2 + 5\gamma'\right),$$

and

$$u_0(1, \theta) = \frac{5\phi(2 + \gamma')\sin(2\theta)}{2},$$

thereby recovering the results of Style et al.\textsuperscript{35} Interestingly, we find that in this limit, at the exact cloaking point ($\gamma' = 2/3$), the droplet will stretch less than the host material $\text{viz.}, (l - 2R)/(2R) = (5/8)\phi_{\text{rel}}$. However, the droplet will stretch the same amount as the host material at $\gamma' = 4/15 < 2/3$, within the softening regime. The predictions of Eshelby’s theory\textsuperscript{42} and effective droplet strain (10/3)$\phi_{\text{rel}}$ are recovered for $\phi, \gamma' \ll 1$, whereas an unperturbed spherical shape is found when $\gamma' \gg 1$, for arbitrary values of $\phi$.

Finally, in Fig. 4, we compare $E_{\text{eff}}$ for this theory (red) with a modified version of the Mori–Tanaka theory (green\textsuperscript{33}) and the dilute theory (blue\textsuperscript{35}). We see that this theory predicts a more pronounced softening in the softening regime ($\gamma' < 2/3$), and a more pronounced stiffening in the stiffening regime ($\gamma' > 2/3$) than does the modified Mori–Tanaka theory. Interestingly, in the stiffening regime, the three-phase and dilute theories are perhaps experimentally indistinguishable, well beyond the concentration range where the latter is expected break down ($\phi \approx 0.2$). This indicates that, depending on the range of $\gamma'$ of relevance, the dilute theory provides a simple framework for comparison with experiment given that it is the appropriate asymptotic limit of the non-dilute theory. All three theories predict exactly the same mechanical cloaking condition, $\gamma' = 2/3$, of the inclusions, independent of $\phi$. We note here that the results in the surface-tension free limit $\gamma' \to 0$ are compared with the classical result\textsuperscript{12} in Appendix A.

### III. Conclusions

Based on a three-phase generalized self-consistent approach, we have estimated elastic moduli of composites including liquid droplets by taking into account the (linear-elastic) solid/droplet interfacial surface tension. In the limit $\phi \to 0$, we recover the dilute-theory expressions of Style et al.\textsuperscript{35,36} The Young’s modulus of the composite depends on $\gamma'$, which is the ratio of the elastocapillary length $L$, to the inclusion radius $R$. The results are compared quantitatively to the dilute theory and a version of Mori–Tanaka theory, both of which include surface tension. In the softening case, the three-phase theory and the modified Mori–Tanaka theory are consistent over a wide range of $\gamma'$, whereas in the stiffening case the three-phase theory is consistent with the dilute theory even for volume fractions over which the latter is expected to break down. All three models predict cloaking of the far-field effects associated with the inclusions when $R = 3L/2$ or $\gamma' = 2/3$ for all volume fractions $\phi$. We have calculated both the effective droplet strain and radial and polar interfacial displacements and found that, in the dilute limit and at exact cloaking, the droplet strain is smaller than that of the host material, whereas they are equal at $\gamma' = 4/15 < 2/3$, within the softening regime.

Finally, we note that there is an interesting similarity between the mechanical response of the multi-phase soft materials studied here and what one finds in poroelasticity, which is a framework used to study the effective medium response of fluid filled host structures, applied to problems ranging from biology to geophysics [e.g., ref. 43–48]. For example, in many biological settings, the composite medium has soft elastic or liquid inclusions, and the deformation of the host material is controlled by the value of $\phi$, which is typically determined as part of the solution to the problem. Whereas in poroelasticity a major challenge involves the modeling of the flow permeability, which is specified as a function of $\phi$, our approach derives the mechanical response as a function of $\phi$. We suggest that by treating the mechanical properties of poroelastic media within the framework studied here, one may be able to constrain the $\phi$ dependence of transport properties such as the flow permeability.

### Appendix A: coefficients in eqn (15)

The coefficients in eqn (15) are

\[
\begin{align*}
    a_0 &= -49 - 252\phi^2 + 25\nu_2^2 - 25\phi^3(7 + \nu_2^2) - 25\phi^7(-7 + \nu_2^2) + 2\phi^2(-49 + 25\nu_2^2) \\
    a_1 &= -7 + 504\phi^2 + 30\nu_2 + 150\phi^3(-3 + \nu_2)\nu_2 + 25\phi^5(7 + \nu_2^2) - 3\phi^4(49 - 140\nu_2 + 75\nu_2^2) \\
    a_2 &= 56 - 252\phi^2 - 30\nu_2 - 50\phi^2 - 25\phi^7(-7 + \nu_2^2) + 50\phi^7(7 - 12\nu_2 + 8\nu_2^2) + 4\phi^7(49 - 105\nu_2 + 50\nu_2^2)
\end{align*}
\]

and

\[
\begin{align*}
    b_0 &= 252\phi^2(1 + 2\nu_2) + 25\phi^7(-7 + \nu_2^2) - 50\phi^7(-7 + 6\nu_2 + 4\nu_2^2) + 4(49 - 63\nu_2 + 20\nu_2^2) + 2\phi^7(-119 + 48\nu_2 + 95\nu_2^2) \\
    b_1 &= -150\phi^2(-3 + \nu_2)\nu_2 - 504\phi^2(-1 + 2\nu_2) + 100\phi^3(-7 + 6\nu_2 + 4\nu_2^2) + 2(7 - 39\nu_2 + 20\nu_2^2) - 3\phi^4(119 - 388\nu_2 + 285\nu_2^2) \\
    b_2 &= 2(126\phi^2(-1 + 2\nu_2) - 25\phi^7(-7 + 6\nu_2 + 4\nu_2^2) - 25\phi^7(7 - 12\nu_2 + 8\nu_2^2) - 4(28 - 51\nu_2 + 20\nu_2^2) + 2\phi^7(238 - 606\nu_2 + 380\nu_2^2)
\end{align*}
\]
We note here that eqn (3.14) of Christensen and Lo\textsuperscript{12} is incorrect in the regime of large droplets ($R \gg L$). In this regime, the condition (15) reduces to $a_0 + a_1 \mu_{rel} + a_2 \mu_{rel}^2 = 0$, whose solution $\mu_{rel} = \mu_{rel,RL}[\phi, \mu_2]$ is invariant under $\mu_2$-scalings, as expected from the symmetries of the equations of the elastostatics.

**Appendix B: shape of the droplets under uniaxial stress**

By using our model, we determine the shape of the generic droplet embedded in an incompressible matrix ($v_2 = 1/2$) undergoing uniaxial stress. Note that, with $\varepsilon_{rel}^0 = \varepsilon_{rel}^0/2$, the purely deviatoric far-field strain conditions of eqn (2) are equivalent to the strain system of Style et al., [ref. 35, see 3 lines below eqn (2)]. Substituting the expressions for $A_2 - G_2$ into eqn (3) and (4), we obtain the following surface displacements,

\[
\frac{u_x(1, \theta)}{R} = [6A_2 + 2B_2 + 6C_2 - 3D_2] \varphi_2(\cos \theta), \quad (B1)
\]

and

\[
\frac{u_y(1, \theta)}{R} = [5A_2 + B_2 + D_2] \frac{d \varphi_2(\cos \theta)}{d \theta}. \quad (B2)
\]

Finally, we note that the coefficients $[f_i]$ in eqn (16) and (17) are:

\[
\begin{align*}
    f_1 &= 40(\mu_{rel} - 1) - 21(\mu_{rel} - 1) x^2 + (19 + 16 \mu_{rel}) x^7 \\
    f_2 &= 6 \left(38(\mu_{rel} - 1)^2 + 75(\mu_{rel} - 1)(2 + 3 \mu_{rel}) x^2 + 112(2 + \mu_{rel} - 3 \mu_{rel}) x^7 + 50(\mu_{rel} - 1)(3 + 4 \mu_{rel}) x^2 + (2 + 3 \mu_{rel})(19 + 16 \mu_{rel}) x^{10} \right) \\
    f_3 &= 15 \left(-48(\mu_{rel} - 1)^2 + 40(\mu_{rel} - 1)(2 + 3 \mu_{rel}) x^2 + 30(3 + \mu_{rel} - 4 \mu_{rel}) x^7 + (2 + 3 \mu_{rel})(19 + 16 \mu_{rel}) x^{10} \right) \\
    f_4 &= 10(\mu_{rel} - 1) + 28(\mu_{rel} - 1) x^2 + 2(19 + 16 \mu_{rel}) x^7 \\
    f_5 &= 3(40(\mu_{rel} - 1) - 56(\mu_{rel} - 1) x^2 + (19 + 16 \mu_{rel}) x^7)
\end{align*}
\]

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**References**

10. See also e.g. ref. 12 or ref. 49 and 50, although unjustified assumptions are invoked in these two latter papers. Note further that the relative effective shear modulus formula $\mu/\mu_m$ presented in ref. 12 (eqn (3.14) through (3.18)) should be revised. This is evident from its symmetry breaking property under arbitrary rescaling of the matrix shear modulus $\mu_m$, whereas $\mu/\mu_m$ is expected to be invariant in the case of (interface stress-free) liquid inclusions. Such a symmetry breaking is clear as $n_2$ in their eqn (3.18), and hence also the solution $\mu/\mu_m$ of the quadratic eqn (3.14), is $\mu_m$-dependent even in the liquid inclusion limit $\mu_i \to 0$.
51 Note that we could well have chosen $\varepsilon_d = \left(\frac{l - 2R}{2R}\right)$ rather than $\left(\frac{l - 2R}{2R}\right)$, but chose the latter to facilitate comparison to the dilute results from ref. 35.