Hashin's bounds for elastic properties of particle-reinforced composites with graded interphase

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Abstract

The paper is focused on analytical prediction of the effective bulk and shear modulus for particulate composites reinforced with solid spherical particles surrounded by graded interphase zone. A three-dimensional elasticity problem for a single inclusion embedded in a finite matrix is studied. The graded interphase zone around the inclusion is assumed to have power law variation of the shear modulus with radial co-ordinate, with Poisson's

ratio assumed to be constant and equal to that of the matrix. Following Hashin's approach, two boundary value problems are considered and stress and displacement fields in the interphase zone are determined. They are then used to calculate the elastic energy for the single inclusion composite under spherically symmetric state and pure shear state and derive closed-form expressions for the bulk modulus and the upper and lower bounds for the shear modulus. Numerical results for hard and soft interphase zones are presented and discussed for a range of the interphase zone thickness ratios.

Keywords: Particle reinforced composites; Elastic moduli; Spherical inclusions; Interphase effects.

1 Introduction

Macroscopic properties of particle-reinforced composite materials are strongly influenced by the phenomena at the interface between particles and the matrix. For example, study of finished and unfinished graphite fibres in epoxy matrices (Drzal et al, 1983) revealed that the finish layer, i.e. an epoxy-compatible coating applied to fibres with the view to enhance their adhesion with the matrix, creates a brittle interphase layer between the fibre and matrix which increases the interfacial shear strength but at the expense of changing the failure mode from interfacial to matrix.

To describe the effect of interfacial phenomena on composites properties, either an imperfect interface is considered (see e.g., Lipton & Talbot 2001; McArthur & Sudak 2016), or an interphase zone between particles and the matrix is introduced, with properties that differ from those of both main phases (see e.g., Voros & Pukanszky 2001; Duan et al. 2005; Bienveniste & Baum 2007).

For composites reinforced with spherical particles, Hashin (1991) proposed to model an imperfect interface between particles and the matrix as a thin interphase zone consisting of a single homogeneous layer, with properties that are different from the properties of particles and the matrix. He analysed the resulting three-phase composite material using the compos-

ite spheres assemblage and the generalised self-consistent scheme models and investigated the interphase effect on the effective bulk and shear modulus and the thermal expansion coefficient. Later, Hashin & Monteiro (2002) used the three-phase model of particle-reinforced composite to inversely determine the interphase zone properties from the experimentally measured properties of the composite using the generalised self-consistent scheme.

Hervé & Zaoui (1993, 1995) developed a micromechanical model for composites reinforced with spherical particles surrounded by multi-layered coatings/interphases with homogeneous layers. They replaced the inhomogeneous inclusion (comprising the particle and multi-layered coating/interphase) with an equivalent homogeneous inclusion and went on to predict the bulk and shear modulus of the composite.

Approximation of radially varying properties of the interfacial transition zone by multiple concentric layers with piecewise-constant properties was explored by Garboczi and Bentz (1997) and Garboczi and Berryman (2000) as applied to concrete. For small volume fractions of aggregate, analytical formula was derived for the bulk modulus and thermal expansion coefficient.

Experimental results for polymeric materials and concrete indicate that properties of the interphase zone are not uniform through its thickness but vary radially outward from the centre of the inclusion (see e.g. Holliday & Robinson 1973; Lutz et al. 1997). On the basis of these observations, a number of researchers have assumed specific profiles for the variation of properties in the interphase zone, which then enabled them to predict the mechanical properties of particulate composites using a variety of methods.

Lutz & Zimmerman (1996, 2005) modelled graded interphase around the inclusion as graded matrix, with power law variation of elastic properties allowing a smooth transition between the interphase and the matrix. They used the method of Frobenius series to derive an expression for the effective elastic moduli of a material with a dispersion of inclusions. The model was successfully used by Lutz et al. (1997) to predict the bulk modulus of concrete. A similar approach was used for thermal/electrical conductivity. In the graded interphase model of Lutz & Zimmerman (1996, 2005), the thickness of interphase zone is not specified

but can be set according to a chosen criterion. By using these models, Sburlati & Cianci (2015) determined the bulk modulus expression in terms of hypergeometric functions for hollow and solid inclusions and Sburlati & Monetto (2016) performed a parametric investigations on bulk modulus. In similar way, in Sburlati et al. (2017), the coefficient of thermal expansion was determined.

Wang & Jasiuk (1998) considered a composite with spherical inclusions and represented the interphase as a functionally graded material of finite thickness, with power law variation of the Young's modulus and constant Poisson's ratio, or with both Young's modulus and Poisson's ratio varying linearly or cubically through the thickness. They calculated the effective bulk modulus using the composites spheres assemblage method and the effective shear modulus using the generalised self-consistent method.

Shen & Li (2003, 2005) proposed an effective interphase model and a uniform replacement method to study the effect of an inhomogeneous interphase with varying elastic properties in the radial direction on the effective elastic moduli of composites reinforced by spherical particles. Using a modified technique of Shen & Li (2003, 2005), Sevostianov & Kachanov (2006, 2007) investigated the effect of graded interface on the elastic moduli, conductive and thermal properties of particulate nanocomposites. The interphase was treated as a layer of finite thickness with elastic moduli that smoothly vary from a set minimum value to the moduli of the matrix. The authors concluded that the effect of the matrix/inclusion interface is controlled mainly by the interphase thickness and less so by the particular profile of property variation as long as it is changes rapidly and levels smoothly toward the matrix.

Andrianov et al. (2010) performed an asymptotic analysis of imperfect interfaces in the conduction problem for particle-reinforced composites. Imperfect interfaces were treated as thin homogeneous layers surrounding the particles, with distinct properties and thickness, which was made to approach zero to develop a solution. The influence of the interface properties on the effective conductivity and on the local potential and flux fields was investigated.

Comparative analysis of different approaches to modelling imperfect interfaces in fibre-reinforced composites was performed by Sevostianov et al (2012). It was concluded that if

the contrast between fibre and matrix properties is large, there is little difference in effective elastic properties of the composite as predicted by the differential approach, three-phase model and spring model.

Nazarenko et al. (2016) proposed a new approach to the determination of equivalent inhomogeneity for spherical particles and the spring layer model of their interphases with the matrix material, suitable for thin compliant interphases where displacement jumps are significant but stress jumps are small. The properties of equivalent inhomogeneity, incorporating only properties of the original inhomogeneity and its interphase, are determined employing a new approach based on the exact Lurié's solution for spheres.

Focussing on fibre-reinforced composites, Andrianov et al (2017) considered an infinitely thin interface on the phase boundary, the properties of which are the average value of the properties of the matrix and fibres. This interface model model was used to derive the effective asymptotic formulae for conductivity of densely packed fibre-reinforced composites, including the case of non-conducting fibres contacting each other through a thin conducting interface.

The aim of this paper is to predict analytically bounds for the effective bulk and shear modulus of particulate composites reinforced with solid spherical particles surrounded by graded interphase zone, using the composite spheres assemblage method of Hashin (1960, 1962) and Hashin & Shtrikman (1963). The paper is organised as follows. In Section 2, three-dimensional elasticity problems for a single inclusion embedded in a finite matrix are formulated. The graded interphase zone around the inclusion is assumed to have power law variation of the shear modulus with radial co-ordinate, and Poisson's ratio is assumed to be constant and equal to that of the matrix. In Section 3, explicit solutions for spherically symmetric condition and shear condition respectively with displacement and traction boundary conditions are developed and stress and displacement fields are determined. In Section 4, they are used to calculate the elastic energy for the single inclusion composite under radially symmetric condition while pure shear state and derive closed-form expressions for the bulk modulus and the upper and lower bounds for the shear modulus. In Section 5, numerical

results for hard and soft interphase zones are presented and discussed for a range of the interphase zone thickness ratios. The effect of Poisson's ratio of the graded interphase zone on the bulk modulus value is also investigated and discussed.

2 Problem formulation

Consider a composite material with solid spherical inclusions embedded in an isotropic matrix, with a non-homogeneous interphase zone between each inclusion and the matrix. The volume fraction of the inclusions with the interphase zone is assumed to be uniform throughout the composite. On a macroscopic scale, the composite material is assumed to be quasi-homogeneous and quasi-isotropic.

In order to determine the shear and bulk modulus of the above composite, we use the composite spheres assemblage model (CSA) of Hashin (1960, 1962) in which a spherical representative volume element of radius R containing one inclusion is adopted.

The element is referred to spherical co-ordinate system $(0; r, \theta, \phi)$ (Fig.1) and consists so of the representative sphere of radius R, concentric with a solid spherical inclusion of radius b, and a non-homogeneous interphase zone $(b \leq r \leq c)$ surrounding the inclusion $(c \leq R)$. We assume that the matrix is isotropic and homogeneous, with the shear modulus μ_m and Poisson's ratio ν_m, and the inclusion $(0 \leq r \leq b)$, is also isotropic and homogeneous, with elastic properties μ_i and ν_i.

We assume that the shear modulus of the non-homogeneous interphase zone varies in the radial direction according to the power law in the form

$$\mu(r) = \mu_m \left(\frac{c}{r}\right)^\beta \quad \text{with} \quad \mu(b) = \mu_{ip}. \tag{2.1}$$

At its interface with the matrix $(r = c)$, the graded interphase zone has the same value of the shear modulus μ_m as the matrix. The inhomogeneity parameter β controls the profile of the power law in the interphase zone and can be determined as

$$\beta = \frac{\ln(\mu_{ip}) - \ln(\mu_m)}{\ln(c) - \ln(b)}. \tag{2.2}$$

Several advantageous aspects of the present interface model are worth mentioning. The model imposes no restrictions on the interface thickness, as there is no requirement that the interface should be a thin layer. The model can be applied to both harder-than-matrix and softer-than-matrix interphase zones. This is not the case for other interface models, for example those where the properties of the interface are taken as the average value of matrix and inclusion properties. The graded interphase zone is treated as a single inhomogeneous layer rather than a set of multiple homogeneous layers, with properties varying as a continuous function of radial coordinate. The properties of the graded interphase zone depend on the properties of the matrix (they match properties of the matrix at the outer boundary of the graded interphase zone) and are independent from the properties of inclusions. The profile of the interphase zone can be controlled via the inhomogeneity parameter.

We assume that Poisson's ratio of the interphase zone has the same value ν_m as the Poisson's ratio of the matrix. This assumption is not as restrictive as it may seem, since the value of Poisson's ratio seems to have negligible effect on the shear modulus bounds, similarly to the case of a particle-reinforced composite without the interphase.

We assume that perfect bonding exists at all interfaces; therefore, the following continuity conditions for stresses and displacements are fulfilled:

$$[\sigma_r]_{r=b} = 0, \quad [\sigma_{r\theta}]_{r=b} = 0, \quad [\sigma_{r\phi}]_{r=b} = 0, \quad [u_r]_{r=b} = 0, \quad [u_\theta]_{r=b} = 0, \quad [u_\phi]_{r=b} = 0,$$

$$[\sigma_r]_{r=c} = 0, \quad [\sigma_{r\theta}]_{r=c} = 0, \quad [\sigma_{r\phi}]_{r=c} = 0, \quad [u_r]_{r=c} = 0, \quad [u_\theta]_{r=c} = 0, \quad [u_\phi]_{r=c} = 0.$$

$$(2.3)$$

In order to determine the bulk and shear modulus of the composite sphere shown in Fig.1, we adopt Hashin's energy approach (1962), and consider two different boundary value problems that lead, respectively, to the effective bulk modulus and to the upper and lower bounds for the shear modulus. In this way, we adopt, in $r = R$, radially symmetric boundary conditions to determine bulk modulus and shear boundary conditions in the plane $z = 0$ to determine the shear bounds.

2.1 Spherically symmetric boundary conditions

In order to obtain the bulk modulus for the problem shown in Fig.1, spherically symmetric problems are considered assuming at $r = R$ the following two conditions.

- *Displacement boundary value problem*

$$u^{(m)}(R) = s\,R, \tag{2.4}$$

   where s is the normal strain.

- *Traction boundary value problem*

$$\sigma_r^{(m)}(R) = 3\,K_m\,s, \tag{2.5}$$

   where K_m is the bulk modulus of the matrix.

2.2 Shear boundary conditions

To obtain the shear modulus for the problem shown in Fig.1, we recall that, for a generic homogeneous sphere (S) of radius R with shear modulus μ in a pure shear state, we have

$$
\begin{aligned}
u_x^{(S)} &= \frac{\gamma}{2}\,y, \qquad u_y^{(S)} = \frac{\gamma}{2}\,x, \qquad u_z^{(S)} = 0, \\
\sigma_{xx}^{(S)} &= 0, \ \ \sigma_{yy}^{(S)} = 0, \ \ \sigma_{xy}^{(S)} = \tau, \ \ \sigma_{xz}^{(S)} = 0, \ \ \sigma_{yz}^{(S)} = 0, \ \ \sigma_{zz}^{(S)} = 0,
\end{aligned}
\tag{2.6}
$$

where $\tau = \mu\,\gamma$.

The displacement and stress components in spherical co-ordinates are

$$
\begin{aligned}
u_r^{(S)}(r,\theta,\phi) &= \frac{\gamma}{2}\,r\sin^2\theta\sin 2\phi, \\
u_\theta^{(S)}(r,\theta,\phi) &= \frac{\gamma}{4}\,r\sin 2\theta\sin 2\phi, \\
u_\phi^{(S)}(r,\theta,\phi) &= \frac{\gamma}{2}\,r\sin\theta\cos 2\phi,
\end{aligned}
\tag{2.7}
$$

and

$$
\begin{aligned}
\sigma_r^{(S)}(r,\theta,\phi) &= \tau\sin^2\theta\,\sin 2\phi, & \sigma_\theta^{(S)}(r,\theta,\phi) &= \tau\cos^2\theta\,\sin 2\phi, \\
\sigma_{r\theta}^{(S)}(r,\theta,\phi) &= \frac{\tau}{2}\,\sin 2\theta\sin 2\phi, & \sigma_{\theta\phi}^{(S)}(r,\theta,\phi) &= \tau\,\cos\theta\cos 2\phi, \\
\sigma_{r\phi}^{(S)}(r,\theta,\phi) &= \tau\,\sin\theta\cos 2\phi, & \sigma_\phi^{(S)}(r,\theta,\phi) &= -\tau\,\sin 2\phi.
\end{aligned}
\tag{2.8}
$$

In this way, we assume the two different cases.

- *Displacement boundary value problem*

  We assume that at the outer boundary the displacement field is given by equation (2.7) with $r = R$ and the material properties are those of the matrix:

$$u_r^{(m)}(R, \theta, \phi) = \frac{\gamma}{2} R \sin^2 \theta \sin 2\phi,$$
$$u_\theta^{(m)}(R, \theta, \phi) = \frac{\gamma}{4} R \sin 2\theta \sin 2\phi, \qquad (2.9)$$
$$u_\phi^{(m)}(R, \theta, \phi) = \frac{\gamma}{2} R \sin \theta \cos 2\phi.$$

- *Traction boundary value problem*

  We assume that, for $r = R$, the stresses are given by equation (2.8) and $\mu = \mu_m$; so we have:

$$\sigma_r^{(m)}(R, \theta, \phi) = \tau \sin^2 \theta \sin 2\phi,$$
$$\sigma_{r\theta}^{(m)}(R, \theta, \phi) = \frac{\tau}{2} \sin 2\theta \sin 2\phi, \qquad (2.10)$$
$$\sigma_{r\phi}^{(m)}(R, \theta, \phi) = \tau \sin \theta \cos 2\phi.$$

3 Elastic solutions

First, we explicitly find elastic solutions for the non-homogeneous interphase zone, while adopting classic solutions available in the literature for the homogeneous matrix and inclusion. In particular, we study the elastic solutions for the spherical symmetry problem and the shear problem in the following two subsections.

3.1 Spherical symmetry solution

The Navier equation for the interphase zone with spherical symmetry and elastic properties described by (2.1) becomes

$$\frac{d^2 u(r)}{dr^2} - \frac{(\beta - 2)}{r} \frac{du(r)}{dr} + \frac{2\left((\beta - 1)\nu_m + 1\right)}{(\nu_m - 1)} \frac{u(r)}{r^2} = 0, \qquad (3.1)$$

in terms of the radial displacement component $u(r)$. See also Hervé and Zaoui (1993) where, in the equilibrium equation at p.2, with our assumption (2.1) we have $\mu'(r)\, r = -\mu(r)\beta$.

The solution of equation (3.1) is

$$u(r) = B_1 r^{h_1} + B_2 r^{h_2}, \qquad (3.2)$$

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where

$$h_1 = \xi + \frac{\sqrt{\xi^2 + (4 - \xi)\,\xi\,\nu_m + 2}}{\sqrt{1 - \nu_m}},$$

(3.3)

$$h_2 = \xi - \frac{\sqrt{\xi^2 + (4 - \xi)\,\xi\,\nu_m + 2}}{\sqrt{1 - \nu_m}},$$

where B_1, B_2 are two integration constants. The stresses components are

$$\sigma_r\,(r) = f_{11}\,B_1 r^{\,h_1 - \beta - 1} + f_{12}\,B_2 r^{\,h_2 - \beta - 1},$$

(3.4)

$$\sigma_\theta\,(r) = \sigma_\phi\,(r) = f_{21}\,B_1 r^{\,h_1 - \beta - 1} + f_{22}\,B_2 r^{\,h_2 - \beta - 1},$$

where

$$f_{11} = \frac{2\,\mu_m c^\beta\,(h_1\,(\nu_m - 1) - 2\,\nu_m)}{2\,\nu_m - 1}, \qquad f_{12} = \frac{2\,\mu_m c^\beta\,(h_2\,(\nu_m - 1) - 2\,\nu_m)}{2\,\nu_m - 1},$$

$$f_{21} = -\frac{2\,\mu_m c^\beta\,(\nu_m h_1 + 1)}{2\,\nu_m - 1}, \qquad f_{22} = -\frac{2\,\mu_m c^\beta\,(\nu_m h_2 + 1)}{2\,\nu_m - 1}.$$

(3.5)

Then, we use the solution for the homogeneous matrix (m) as

$$u^{(m)}\,(r) = A_1 r + \frac{A_2}{r^2},$$

$$\sigma_r^{(m)}\,(r) = -\frac{2\,\mu_m\,(\nu_m + 1)\,A_1}{2\,\nu_m - 1} - \frac{4\,\mu_m A_2}{r^3},$$

(3.6)

$$\sigma_\theta^{(m)}\,(r) = \sigma_\phi^{(m)}\,(r) = -\frac{2\,\mu_m\,(\nu_m + 1)\,A_1}{2\,\nu_m - 1} + \frac{2\,\mu_m A_2}{r^3},$$

and, for the solid homogeneous inclusion (i), as

$$u^{(i)}\,(r) = C_1 r,$$

$$\sigma_r^{(i)}\,(r) = -\frac{2\,\mu_i\,(\nu_i + 1)\,C_1}{2\,\nu_i - 1},$$

(3.7)

$$\sigma_\theta^{(i)}\,(r) = \sigma_\phi^{(i)}\,(r) = -\frac{2\,\mu_i\,(\nu_i + 1)\,C_1}{2\,\nu_i - 1}.$$

We observe that the homogeneous solutions are also obtained for $\beta = 0$, $h_1 = 1$ and $h_2 = -2$ in Eqs.(3.2,4) with the elastic properties of the specific layer.

The five unknown integration constants can be obtained from the continuity conditions (2.3) and boundary conditions at $r = R$ in the displacement form (2.4) or in the traction form (2.5). In Appendix 1 we explicitly write the equation system to obtain the integration constants.

3.2 Shear solution

Following Christensen (2005), we assume the displacement field in the interphase zone in the following form

$$u_r\left(r,\theta,\phi\right) = \frac{1}{2}\,U_r\left(r\right)\sin^2\theta\sin 2\phi,$$

$$u_\theta\left(r,\theta,\phi\right) = \frac{1}{4}\,U_\theta\left(r\right)\sin 2\theta\sin 2\phi, \tag{3.8}$$

$$u_\phi\left(r,\theta,\phi\right) = -\frac{1}{2}\,U_\phi\left(r\right)\sin\theta\cos 2\phi.$$

In this way, the stresses become

$$\sigma_r\left(r,\theta,\phi\right) = \frac{\mu\left(r\right)}{r\left(2\,\nu_m-1\right)}\left(\frac{dU_r\left(r\right)}{dr}\left(\nu_m-1\right)r - 2\,\nu_m\,U_r\left(r\right)\right)\sin^2\theta\sin 2\,\phi+$$

$$+\frac{\nu_m\,\mu\left(r\right)U_\theta\left(r\right)\left(\sin^2\theta - 2\,\cos^2\theta\right)\sin 2\,\phi}{r\left(2\,\nu_m-1\right)} - \frac{2\nu_m\,\mu\left(r\right)\,U_\phi\left(r\right)\sin 2\,\phi}{r\left(2\,\nu_m-1\right)},$$

$$\sigma_\theta\left(r,\theta,\phi\right) = -\frac{\mu\left(r\right)}{r\left(2\,\nu_m-1\right)}\left(\frac{dU_r\left(r\right)}{dr}\nu_m\,r + U_r\left(r\right)\right)\sin^2\theta\sin 2\,\phi+$$

$$-\frac{\mu\left(r\right)U_\theta\left(r\right)\left(\left(\nu_m-1\right)\sin^2\theta + \cos^2\theta\right)\sin 2\,\phi}{r\left(2\,\nu_m-1\right)} - \frac{2\,\nu_m\,\mu\left(r\right)\,U_\phi\left(r\right)\sin 2\,\phi}{r\left(2\,\nu_m-1\right)},$$

$$\sigma_\phi\left(r,\theta,\phi\right) = \sigma_\theta\left(r,\theta,\phi\right) + \frac{\mu\left(r\right)}{r}\left(U_\theta\left(r\right)\sin^2\theta + 2\,U_\phi\left(r\right)\right)\sin 2\,\phi,$$

$$\sigma_{r\theta}\left(r,\theta,\phi\right) = \frac{\mu\left(r\right)}{4}\left(\frac{dU_\theta\left(r\right)}{dr} - \frac{U_\theta\left(r\right)}{r} + 2\,\frac{U_r\left(r\right)}{r}\right)\sin 2\,\theta\sin 2\,\phi,$$

$$\sigma_{r\phi}\left(r,\theta,\phi\right) = -\frac{\mu\left(r\right)}{2}\left(\frac{dU_\phi\left(r\right)}{dr} - \frac{U_\phi\left(r\right)}{r} - 2\,\frac{U_r\left(r\right)}{r}\right)\cos 2\,\phi\sin\theta,$$

$$\sigma_{\theta\phi}\left(r,\theta,\phi\right) = \frac{\mu\left(r\right)}{r}\,U_\theta\left(r\right)\cos\theta\cos 2\,\phi. \tag{3.9}$$

The functions $U_r\left(r\right)$, $U_\theta\left(r\right)$ and $U_\phi\left(r\right)$ are determined from the following set of Navier equations

$$\frac{d^2U_r\left(r\right)}{dr^2} - \left(\beta - 2\right)\frac{1}{r}\frac{dU_r\left(r\right)}{dr} + \frac{3}{2\left(\nu_m-1\right)}\frac{1}{r}\frac{dU_\theta\left(r\right)}{dr}+$$

$$+\frac{5 + 2\left(\beta - 4\right)\nu_m}{\nu_m-1}\frac{U_r\left(r\right)}{r^2} - \frac{9 + 6\left(\beta - 2\right)\nu_m}{2\left(\nu_m-1\right)}\frac{U_\theta\left(r\right)}{r^2} = 0,$$

$$\frac{d^2U_\theta\left(r\right)}{dr^2} - \left(\beta - 2\right)\frac{1}{r}\frac{dU_\theta\left(r\right)}{dr} - \frac{2}{2\nu_m-1}\frac{1}{r}\frac{dU_r\left(r\right)}{dr}+ \tag{3.10}$$

$$-\frac{2\left(6 - \beta\right)\nu_m + \beta - 12}{2\nu_m-1}\frac{U_\theta\left(r\right)}{r^2} - \frac{4\left(\beta - 2\right)\nu_m - 2\left(\beta - 4\right)}{2\nu_m-1}\frac{U_r\left(r\right)}{r^2} = 0,$$

$$U_\phi\left(r\right) = -U_\theta\left(r\right).$$

The solution of this set of equations, written in terms of four integration constants K_j, $(j = 1, 2, 3, 4)$ is

$$U_r(r) = \left(K_1 r^{\Lambda_2} + K_2 r^{-\Lambda_2} + K_3 r^{\Lambda_1} + K_4 r^{-\Lambda_1}\right) r^\xi,$$

$$U_\theta(r) = -U_\phi(r) = \left(K_1 Q_1 r^{\Lambda_2} + K_2 Q_2 r^{-\Lambda_2} + K_3 Q_3 r^{\Lambda_1} + K_4 Q_4 r^{-\Lambda_1}\right) r^\xi,$$

(3.11)

where we have set $\xi = (\beta - 1)/2$,

$$\Lambda_1 = \frac{1}{2} \sqrt{\frac{2\left(2\left(1 - \nu_m\right) \xi^2 + 2\left(3\nu_m - 1\right) \xi - \left(11\nu_m - 13\right)\right) - \sqrt{\delta}}{\left(1 - \nu_m\right)}},$$

$$\Lambda_2 = \frac{1}{2} \sqrt{\frac{2\left(2\left(1 - \nu_m\right) \xi^2 + 2\left(3\nu_m - 1\right) \xi - \left(11\nu_m - 13\right)\right) + \sqrt{\delta}}{\left(1 - \nu_m\right)}},$$

(3.12)

$$\delta = 16\left(\left(25\nu_m - 22\right)\nu_m + 1\right)\xi^2 - 16\left(5\nu_m - 3\right)\left(5\nu_m - 7\right)\xi + 4\left(25\nu_m - 78\right)\nu_m + 228,$$

(3.13)

and

$$Q_1 = -\left(\left(4\nu_m - 1\right)\xi + \Lambda_2 - 2\nu_m + 3\right)\frac{Q_{12}}{\bar{Q}},$$

$$Q_2 = -\left(\left(4\nu_m - 1\right)\xi - \Lambda_2 - 2\nu_m + 3\right)\frac{Q_{12}}{\bar{Q}},$$

$$Q_{12} = \left(2\nu_m - 1\right)\left(\left(\nu_m - 1\right)\xi^2 - 4\nu_m\xi - \left(\nu_m - 1\right)\Lambda_2{}^2\right) + 12\nu_m{}^2 - 16\nu_m + 2,$$

$$Q_3 = -\left(\left(4\nu_m - 1\right)\xi + \Lambda_1 - 2\nu_m + 3\right)\frac{Q_{34}}{\bar{Q}},$$

$$Q_4 = -\left(\left(4\nu_m - 1\right)\xi - \Lambda_1 - 2\nu_m + 3\right)\frac{Q_{34}}{\bar{Q}},$$

(3.14)

$$Q_{34} = \left(2\nu_m - 1\right)\left(\left(\nu_m - 1\right)\xi^2 - 4\nu_m\xi - \left(\nu_m - 1\right)\Lambda_1{}^2\right) + 12\nu_m{}^2 - 16\nu_m + 2,$$

$$\bar{Q} = 12\nu_m\left(2\nu_m - 1\right)^2 \xi^2 - 6\left(2\nu_m - 1\right)\left(4\nu_m{}^2 - 7\nu_m + 1\right)\xi +$$

$$+ 6\nu_m\left(\nu_m - 1\right)\left(2\nu_m - 5\right) + 3.$$

We observe that, for the isotropic homogeneous case, we have: $\Lambda_1 = 3/2$, $\Lambda_2 = 7/2$, $\xi = -1/2$ and the quantities Q_1, Q_2, Q_3 and Q_4, assume the following values:

$$Q_1 = \frac{7 - 4\nu_m}{6\nu_m}, \quad Q_2 = -\frac{2}{3}, \quad Q_3 = 1, \quad Q_4 = \frac{2(2\nu_m - 1)}{4\nu_m - 5}.$$

(3.15)

Now, the functions $U_r^{(m)}, U_\theta^{(m)}$ and $U_\phi^{(m)}$ in the homogeneous matrix (m), obtained by

the solution of (3.9), by putting $\beta = 0$, assume the form

$$U_r^{(m)}\left(r\right) = H_1 r^3 + \frac{H_2}{r^4} + H_3 r + \frac{H_4}{r^2},$$

$$U_\theta^{(m)}\left(r\right) = \frac{7 - 4\,\nu_m}{6\,\nu_m} H_1 r^3 - \frac{2}{3}\frac{H_2}{r^4} + H_3 r + \frac{2\left(2\,\nu_m - 1\right)}{4\,\nu_m - 5}\frac{H_4}{r^2}, \qquad (3.16)$$

$$U_\phi^{(m)}\left(r\right) = -U_\theta^{(m)}\left(r\right),$$

and, in similar way, in the homogeneous solid inclusion (i) we have

$$U_r^{(i)}\left(r\right) = F_1 r^3 + F_3 r,$$

$$U_\theta^{(i)}\left(r\right) = \frac{7 - 4\,\nu_i}{6\,\nu_i} F_1 r^3 + F_3 r, \qquad (3.17)$$

$$U_\phi^{(i)}\left(r\right) = -U_\theta^{(i)}\left(r\right).$$

The unknown integration constants can be obtained from the continuity conditions (2.4) and boundary conditions at $r = R$ in the displacement form (2.8) or in the traction form (2.9). In Appendix 2 we explicitly write the equation system to obtain the integration constants.

4 Application of Hashin's theory

The elastic solutions derived in the previous sections can be used to determine bounds for the elastic moduli of the composite sphere shown in Figure 1.

The general Hashin's theory considers the change in strain energy in a corresponding equivalent homogeneous sphere of radius R and elastic properties K_h and μ_h, due to the presence of nonhomogeneities. The different displacement or traction assumptions at $r = R$ (see section 2) introduced to evaluate the change of the strain energy of the composite sphere of Figure 1 and the equivalent homogeneous sphere, permit us to obtain the bounds for the elastic moduli (Hashin, 1962)).

Before deriving the formulae for these bounds, let us introduce the following quantities:

$$\eta = \left(\frac{b}{R}\right)^3, \qquad \Omega = \frac{c}{b}, \qquad (4.1)$$

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where η is the volume fraction of the inclusions in the composite. We note that, as a consequence of the above definitions and the geometry of the problem (Fig.1), η and Ω are related by the inequality $0 \leq \eta \leq 1/\Omega^3$.

4.1 Bulk modulus

Application of Hashin's method shown that the expressions for the effective bulk modulus K_h obtained with the displacement or traction conditions coincide.

- *Displacement or traction approach*

  We consider the elastic energy U_h stored in a sphere of radius R and volume V, comprised of the effective medium, subjected to boundary conditions (2.4):

  $$U_h = U_0 + \delta U, \tag{4.2}$$

  where

  $$U_0 = \frac{9}{2} V K_m s^2, \tag{4.3}$$

  is the energy stored in an homogeneous sphere of radius R with boundary conditions (2.4) and bulk modulus K_m of the matrix of the composite sphere. Moreover, we define the effective bulk modulus as the value for which it holds

  $$U_h = \frac{9}{2} V K_h s^2. \tag{4.4}$$

  The explicit calculation of the term δU can be done by using Eshelby formula (Eshelby 1951). So doing we get the expression

  $$\frac{K_h}{K_m} = 1 - \frac{N_0 \left(K_m + \frac{4}{3} \mu_m \right) \eta}{N_0 \, K_m \, \eta + 1}, \tag{4.5}$$

where $N_0 = N_1/N_2$ and

$$N_1 = \Omega^{2\,h_2}\,(h_2 - 1)\left(\Omega^\beta\left((3\,K_m + 4\,\mu_m)\,(h_2 - \beta) - 3\,K_m + 8\,\mu_m\right) + 9\,K_i\right)\Omega^4 +$$

$$-\Omega^\beta\,(h_2 - \beta + 2)\left(((3\,K_m + 4\,\mu_m)\,h_2 + 6\,K_m - 4\,\mu_m)\,\Omega^\beta - 9\,K_i\right)\Omega^3,$$

$$N_2 = -\Omega^{2\,h_2}\,(h_2 + 2)\left(\Omega^\beta\left((3\,K_m + 4\,\mu_m)\,(h_2 - \beta) - 3\,K_m + 8\,\mu_m\right) + 9\,K_i\right)K_m\Omega +$$

$$+\Omega^\beta\,(h_2 - \beta - 1)\left(((3\,K_m + 4\,\mu_m)\,h_2 + 6\,K_m - 4\,\mu_m)\,\Omega^\beta - 9\,K_i\right)K_m.$$

$$(4.6)$$

We remark that the case without the interphase is obtained when $h_1 = 1, h_2 = -2, \beta = 0$ and so, the terms N_0 becomes

$$N_0 = \frac{3\,(K_m - K_i)}{(3\,K_i + 4\,\mu_m)\,K_m}. \tag{4.7}$$

4.2 Shear modulus bounds

For the composite shear modulus in the energy approach of Hashin (1960, 1962), the effective shear modulus for the composite material μ_h satisfies the following inequality

$$\mu_h^{(T)} \le \mu_h \le \mu_h^{(u)}, \tag{4.8}$$

where $\mu_h^{(u)}$ and $\mu_h^{(T)}$ are the equivalent shear modulus for the displacement problem and the stress problem, respectively.

- *Displacement approach - upper bound*

  We consider a homogeneous sphere S of radius R, with boundary conditions (2.9) and shear modulus of the matrix of composite sphere μ_m. Then the elastic energy of this sphere is

  $$U_0^{(u)} = \frac{1}{2}\,\mu_m\,\gamma^2\,V, \tag{4.9}$$

  where V is the volume of the sphere S.

  Similarly, for the equivalent homogeneous sphere with $\mu = \mu_h^{(u)}$, and the same boundary conditions (2.10), the elastic energy $U_h^{(u)}$ of this sphere is in the form

  $$U_h^{(u)} = \frac{1}{2}\,\mu_h^{(u)}\,\gamma^2\,V. \tag{4.10}$$

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Following Hashin (1960, 1962) and by using Eshelby's formula (Eshelby 1951), the elastic energy (4.7) can be written as

$$U_h^{(u)} = U_0^{(u)} + \delta U^{(u)}, \tag{4.11}$$

where

$$\delta U^{(u)} = -\frac{4\pi \, \mu_m \, \gamma \, (\nu_m - 1)}{4 \, \nu_m - 5} \, H_4. \tag{4.12}$$

The coefficient H_4 is obtained using displacement boundary value problem.

In this way, we obtain

$$\mu_h^{(u)} = \mu_m + \frac{\delta U^{(u)}}{\frac{1}{2} \, \gamma^2 \, V}. \tag{4.13}$$

In explicit form, the normalized shear modulus $\mu_h^{(u)}$ can be written as

$$\frac{\mu_h^{(u)}}{\mu_m} = 1 + \frac{b_{11} \, \eta^{10/3} + b_{12} \, \eta}{b_{21} \, \eta^{10/3} + b_{22} \, \eta^{7/3} + b_{23} \, \eta^{5/3} + b_{24} \, \eta + b_{25}}, \tag{4.14}$$

where the quantities b_{ij} are

$$b_{11} \, \Omega^{-10} = 30 \, (1 - \nu_m) \, ((2 \, \nu_m - 1) \, (6 \, (19 \, \nu_m - 7) \, \mu_m V_1 - 3 \, (\nu_m - 7) \, V_2 + 27 \, \nu_m V_3)) +$$

$$+ 30 \, (1 - \nu_m) \, ((4 \, (28 - 31 \, \nu_m) \, V_4 + 6 \, (4 \, \nu_m - 7) \, V_5) \, \mu_m + 3 \, (10 \, \nu_m - 7) \, V_6) \,,$$

$$b_{12} \, \Omega^{-3} = 30 \, (3 \, (2 \, \nu_m - 1) \, (2 \, (4 \, V_1 \, \mu_m + V_2) + 3 \, V_3)) \, (\nu_m - 1) \, (10 \, \nu_m - 7) +$$

$$- 2 \, (30 \, \mu_m \, (2 \, V_4 + 3 \, V_5) - 45 \, V_6) \, (\nu_m - 1) \, (10 \, \nu_m - 7) \,,$$

$$b_{21} = -\frac{2 \, (5 \, \nu_m - 4) \, b_{11}}{15 \, (\nu_m - 1)},$$

$$b_{22} \, \Omega^{-7} = 150 \, (1 - 2 \, \nu_m) \, (2 \, (\nu_m \, (\nu_m + 12) - 7) \, \mu_m V_1 + (2 \, \nu_m \, (\nu_m - 3) + 7) \, V_2) +$$

$$- 25 \, (27 \, \nu_m \, (2 \, \nu_m - 1) \, V_3 - (4 \, \nu_m \, (32 \, \nu_m - 57) + 112) \, \mu_m V_4 + 6 \, (7 - 2 \, \nu_m \, (2 \, \nu_m + 3)) \, \mu_m V_5) +$$

$$- 75 \, (4 \, \nu_m \, (2 \, \nu_m - 3) + 7) \, V_6,$$

$$b_{23} \, \Omega^{-5} = 63 \, (2 \, \nu_m - 1) \, (16 \, (\nu_m + 2) \, \mu_m V_1 - 12 \, (\nu_m - 2) \, V_2 + 3 \, (\nu_m + 5) \, V_3) +$$

$$+ 63 \, (4 \, (\nu_m - 3) \, \mu_m V_4 - 12 \, (2\nu_m - 1) \, \mu_m V_5 + 6V_6) \,,$$

$$b_{24} = \frac{5 \, (4 \, \nu_m \, (2 \, \nu_m - 3) + 7) \, b_{12}}{6 \, (1 - \nu_m) \, (10 \, \nu_m - 7)},$$

$$\tag{4.15}$$

$$b_{25} = 2\,(2\,\nu_m - 1)\,(24\,(19\,\nu_m - 17)\,\mu_m V_1 + 6\,(13\,\nu_m - 11)\,V_2 + 9\,(7\,\nu_m - 5)\,V_3)\,(10\,\nu_m - 7) +$$

$$-4\,(\mu_m\,(19\,\nu_m - 17)\,(2\,V_4 + 3\,V_5) - 3\,(5\,\nu_m - 4)\,V_6)\,(10\,\nu_m - 7)\,.$$

$$(4.16)$$

The quantities $V_i, (i = 1...6)$ are given in Appendix 3.

- *Traction approach - lower bound*

  For the traction problem, we consider a homogeneous sphere with $\mu = \mu_m$ with boundary conditions at $r = R$ given by (2.10). Similarly to the previous case, we have

  $$U_0^{(T)} = \frac{\tau^2}{2\,\mu_m}\,V. \tag{4.17}$$

  For the equivalent homogeneous sphere with $\mu = \mu_h^{(T)}$ and boundary conditions at $r = R$ given by (2.11), we have

  $$U_h^{(T)} = \frac{\tau^2}{2\,\mu_h^{(T)}}\,V. \tag{4.18}$$

  In this case the elastic energy (4.11) can be written as

  $$U_h^{(T)} = U_0^{(T)} + \delta U^{(T)}, \tag{4.19}$$

  where

  $$\delta U^{(T)} = \frac{4\pi\,\mu_m\,\tau\,(\nu_m - 1)}{4\nu_m - 5}\,H_4. \tag{4.20}$$

  The coefficient H_4 is obtained from the traction boundary value problem.

  So, we get

  $$\frac{1}{\mu_h^{(T)}} = \frac{1}{\mu_m} + \frac{\delta U^{(T)}}{\frac{1}{2}\,\tau^2\,V}. \tag{4.21}$$

  In explicit form, the normalized shear modulus $\mu_h^{(T)}$ becomes

  $$\frac{\mu_h^{(T)}}{\mu_m} = 1 + \frac{B_{11}\,\eta^{10/3} + B_{12}\,\eta}{B_{21}\,\eta^{10/3} + B_{22}\,\eta^{7/3} + B_{23}\,\eta^{5/3} + B_{24}\,\eta + B_{25}}, \tag{4.22}$$

where the quantities B_{ij} are written in terms of b_{ij} as

$$B_{11} = \frac{4\,(10\,\nu_m - 7)\,b_{11}}{5\,\nu_m + 7}, \quad B_{21} = \frac{4\,(10\,\nu_m - 7)\,b_{21}}{5\,\nu_m + 7},$$

$$B_{22} = \frac{4\,(10\,\nu_m - 7)\,b_{22}}{5\,\nu_m + 7}, \quad B_{23} = \frac{4\,(10\,\nu_m - 7)\,b_{23}}{5\,\nu_m + 7},$$

$$B_{24} = \frac{4\,(\nu_m\,(5\,\nu_m + 3) + 7)\,(10\,\nu_m - 7)\,b_{24}}{5\,(5\,\nu_m + 7)\,(4\,\nu_m\,(2\,\nu_m - 3) + 7)}, \tag{4.23}$$

$$B_{12} = b_{12}, \quad B_{25} = b_{25}.$$

For small volume fraction of the inclusions, the expressions for the upper and lower bounds become the same and reduce to the following expression

$$\frac{\mu_h}{\mu_m} = 1 + \frac{b_{12}}{b_{25}}\,\eta. \tag{4.24}$$

5 Numerical results and discussion

In this section, numerical results for the macroscopic bulk and shear modulus with and without the interphase zone are presented and discussed.

The following elastic properties are considered for the matrix and the solid inclusion: $\mu_i = 3.6\,\mu_m$, $\nu_i = 0.7\,\nu_m$. These values correspond to those used by Hashin (1962).

For the interphase zone, a range of the interphase zone thickness ratios is analysed: $\Omega = 1.1, 1.25, 1.5$. As it was pointed out at the beginning of Section 4, η and Ω are related by the inequality $0 \leq \eta \leq 1/\Omega^3$.

Two cases of elastic properties are analysed: hard interphase with $\mu_{ip} = 1.5\,\mu_m$ and soft interphase with $\mu_{ip} = 0.5\,\mu_m$ and Poisson's ratio in matrix $\nu_m = 0.3$.

Figure 2 shows variation of the shear modulus through the thickness of the graded interphase zone, as given by equation (2.1), for different values of Ω and the hard and soft cases. The values of β for specific interphase zone thickness ratios are determined using expression (2.3).

Figure 3 shows the bulk modulus obtained from the formula (4.2). The red line represents the case without the interphase zone, solid lines represent the hard cases and the dashed

lines represent the soft cases. We observe that the effect of interphase zone on the bulk modulus is the most pronounced in the case of soft interphase zone.

The bulk modulus of a composite with spherical inclusions, in which the interphase zone had two Lamé constants varying in radial direction, was investigated previously by Sburlati & Cianci (2015) and Sburlati & Monetto (2016). Although the power law (2.1) of the present paper is more restrictive compared to the one used in the above-cited papers, this power law allows us to obtain the shear modulus bounds in a simpler way. It is interesting to note that the interphase zone model introduced in this paper assumes Poisson's ratio of the interphase zone to be equal to Poisson's ratio of the matrix. In order to examine the effect of Poisson's ratio variation in the interphase, we compare the bulk modulus prediction obtained by equation (4.2) with that obtained in Sburlati & Cianci (2015).

Figure 4 shows variation of the normalized shear modulus in the interphase zone obtained from equation (2.1) of the present paper and the power law given by equation (2.1) of the paper of Sburlati & Cianci (2015), with the inhomogeneity parameter $\beta = 10$. Both variations are shown for the case of the hard interphase with $\Omega = 1.25$. It is worth pointing out that in the present interphase zone model, the thickness of the zone is defined exactly, whereas in the previous interphase zone model, like in Lutz & Zimmerman (1996, 2005), the thickness of the interphase zone is not defined clearly but instead the whole ma3trix is treated as a graded medium and the interphase zone thickness can be set according to a chosen criterion. Comparison of numerical results produced by the two different power-law variations allows us to examine the effect of the Poisson's ratio on the bulk modulus.

Figure 5 shows comparison of the normalized bulk modulus obtained from equation (4.2) of the present paper and from the expression (5.4) of the paper Sburlati & Cianci (2015). The case without the interphase is shown in red for reference. When Poisson's ratio of the interphase zone is equal to Poisson's ratio of the matrix, $\nu_m = 0.3$, predictions of both models are very close. When Poisson's ratio of the interphase zone is not equal to Poisson's ratio of the matrix, only the previous model is able to predict the bulk modulus. Normalized bulk modulus for different Poisson's ratios of the interphase zone ($\nu_{ip} = 0.27, \nu_{ip} = 0.24$) is

shown in black; $\nu_m = 0.3$, and the value of the shear modulus is assumed to be the same. We observe that as Poisson's ratio of the interphase zone decreases, the bulk modulus of the composite decreases too. Therefore, the assumption of Poisson's ratio of the interphase zone being the same as Poisson's ratio of the matrix, used in the present model, leads to an overestimation of the bulk modulus value. In the following we investigate on shear modulus bounds obtained in subsection 4.2.

Figure 6 shows the upper (continuous lines) and lower (dashed lines) bounds for the normalized shear modulus μ_h/μ_m obtained from the exact expressions (4.11) and (4.18), respectively for hard interphases (a) and soft interphases. The red lines show the bounds for the composite without the interphase zone considered by Hashin (see Figure 5 of Hashin (1962)). They are obtained from the present model by putting ($\Omega = 1$). We observe that composites with and without the interphase zone exhibit similar behaviour for all values of Ω considered. Also, for thin interphase zone, i.e. ($\Omega = 1.1$), the gap between the lower and upper bounds at the maximum value of $\eta = 0.75$ is greater for the soft interphase zone than for the hard interphase zone.

6   Concluding Remarks

The bulk and shear modulus of particulate composites reinforced with solid spherical particles surrounded by the graded interphase zone at the particle/matrix interface have been analysed. Two assumptions about the elastic properties of the graded interphase zone were made: (i) power law variation of the shear modulus with radial co-ordinate and (ii) Poisson's ratio of the interphase zone being equal to that of the matrix (the second assumption is not as restrictive as it may seem, since the value of Poisson's ratio appears to have negligible effect on the shear modulus bounds).

The two assumptions have enable us to determine stress and displacement fields in a spherical representative volume element containing a single particle (inclusion), when either displacement or traction boundary conditions are prescribed at the outer boundary. This

in turn has allowed us, following Hashin's approach, to determine the elastic energy of the single inclusion composite and derive closed-form expression for the bulk modulus and the upper and lower bounds for the shear modulus.

The effect of graded interface zone on the bulk modulus monotonically increases with the particle volume fraction and is more pronounced for the soft interphase zone than for the hard one.

Comparison of two graded interphase zone models, the one presented in this paper, the other in Sburlati & Cianci (2015), shows that when Poisson's ratio of the interphase zone is equal to Poisson's ratio of the matrix, the values of the composite's bulk modulus predicted by the two models are very close, but the present model has the advantage of being simpler. However, the assumption of Poisson's ratio of the interphase zone being the same as Poisson's ratio of the matrix leads to an overestimation of the bulk modulus value.

Analysis of numerical results for hard and soft interphase zones over a range of the zone thickness ratios revealed that the shear modulus bounds for composites with and without the interphase zone generally behave in a similar manner. Hard graded interphase zone makes the composite stiffer in shear and increases the shear modulus bounds of the composite, while soft graded interphase zone makes the composite more compliant in shear, decreasing the shear modulus bounds. When the interphase zone is thin relative to the radius of the particle, the gap between the lower and upper bounds at the maximum permissible particle volume fraction is greater for the soft interphase zone than for the hard interphase zone. Finally, we remark that the numerical results otained for $\mu_h^{(u)}$ and $\mu_h^{(T)}$ give rise to a medium value that is in agreement with the numerical results obtained in Wang & Jasiuk (1998) by using the generalized self-consistent method (Christensen & Lo, 1979).

Appendix

1. Spherically symmetric boundary value problem

The integration constants C_1, B_1, B_2, A_1 and A_2 can be determined by the following system:

$$
\begin{bmatrix}
a_{11} & a_{12} & a_{13} & 0 & 0 \\
a_{21} & a_{22} & a_{23} & 0 & 0 \\
0 & a_{32} & a_{33} & a_{34} & a_{35} \\
0 & a_{42} & a_{43} & a_{44} & a_{45} \\
0 & 0 & 0 & a_{54} & a_{55}
\end{bmatrix}
\begin{bmatrix}
C_1 \\
B_1 \\
B_2 \\
A_1 \\
A_2
\end{bmatrix}
=
\begin{bmatrix}
0 \\
0 \\
0 \\
0 \\
f
\end{bmatrix}
$$

where

$$
a_{11} = \frac{2\,\mu_i\,(1+\nu_i)}{2\,\nu_i - 1}, \quad a_{12} = f_{11}\,b^{\,h_1-\beta-1}, \quad a_{13} = f_{12}\,b^{\,h_2-\beta-1}, \quad a_{21} = b, \quad a_{22} = -b^{\,h_1},
$$

$$
a_{23} = -b^{\,h_2}, \quad a_{32} = f_{11}\,c^{\,h_1-1}, \quad a_{33} = f_{12}\,c^{\,h_2-1}, \quad a_{34} = \frac{2\,\mu_m\,c^{\beta}\,(1+\nu_m)}{2\,\nu_m - 1},
$$

$$
a_{35} = 4\,\mu_m\,c^{\,\beta-3}, \quad a_{42} = c^{\,h_1}, \quad a_{43} = c^{\,h_2}, \quad a_{44} = -c, \quad a_{45} = -c^{-2},
$$

while, respectively for the displacement or the traction boundary condition, the terms of the last line are

$$
a_{54}^{(u)} = R, \quad a_{55}^{(u)} = R^{-2}, \quad f = f^{(u)} = s\,R,
$$

or

$$
a_{54}^{(T)} = -(1+\nu_m), \quad a_{55}^{(T)} = -2\,(2\,\nu_m - 1)R^{-3}, \quad f = f^{(T)} = -(1+\nu_m)\,s.
$$

2. Shear boundary value problem

The continuity conditions (2.3) in the displacements permit us to write the constants F_1, F_3, H_1 and H_2 in term of the constants K_1, K_2, K_3, K_4, H_3 and H_4. Introducing the quantities

$$
k_1 = K_1\,c^{\,\xi-1+\Lambda_2}, \quad k_2 = K_2\,c^{\,\xi-1-\Lambda_2}, \quad k_3 = K_3\,c^{\,\xi-1+\Lambda_1}, \quad k_4 = K_4\,c^{\,\xi-1-\Lambda_1},
$$

we write

$$F_1 = -\frac{6\,\nu_i\,\Omega^{1-\xi}\,\sum_{j=1}^4 \omega_j\,(Q_j - 1)\,k_j}{b^2\,(10\,\nu_i - 7)},$$

$$F_3 = \frac{6\,\nu_i\,\Omega^{1-\xi}\,\sum_{j=1}^4 \omega_j\,(Q_j - 1)\,k_j}{10\,\nu_i - 7} + \Omega^{1-\xi}\,\sum_{j=1}^4 \omega_j k_j,$$

$$H_1 = \frac{2\,\nu_m\,\sum_{j=1}^4 (3\,Q_j + 2)\,k_j}{7b^2\Omega^2} - \frac{10\,\nu_m H_3}{7\,b^2\Omega^2} - \frac{8\,\nu_m\,(5\,\nu_m - 4)\,H_4}{7\,b^5\Omega^5\,(4\,\nu_m - 5)},$$

$$H_2 = -\frac{1}{7}\,\Omega^5 b^5 \sum_{j=1}^4 (2\,\nu_m\,(3\,Q_j + 2) - 7)\,k_j + \frac{1}{7}\,\Omega^5 b^5\,(10\,\nu_m - 7)\,H_3 +$$

$$+\frac{5\,\Omega^2 b^2\,(8\,\nu_m{}^2 - 12\,\nu_m + 7)\,H_4}{28\,\nu_m - 35},$$

where

$$\omega_1 = \Omega^{-\Lambda_2}, \quad \omega_2 = \Omega^{\Lambda_2}, \quad \omega_3 = \Omega^{-\Lambda_1}, \quad \omega_4 = \Omega^{\Lambda_1}.$$

The remaining constants k_1, k_2, k_3, k_4, H_3 and H_4 are obtained using conditions (2.3) on the stresses and the two boundary conditions (2.9) or (2.10). So doing we get the following system

$$\begin{bmatrix} c_{11} & c_{12} & c_{13} & c_{14} & 0 & 0 \\ c_{21} & c_{22} & c_{23} & c_{24} & 0 & 0 \\ c_{31} & c_{32} & c_{33} & c_{34} & c_{35} & c_{36} \\ c_{41} & c_{42} & c_{43} & c_{44} & c_{45} & c_{46} \\ c_{51} & c_{52} & c_{53} & c_{54} & c_{55} & c_{56} \\ c_{61} & c_{62} & c_{63} & c_{64} & c_{65} & c_{66} \end{bmatrix} \begin{bmatrix} k_1 \\ k_2 \\ k_3 \\ k_4 \\ H_3 \\ H_4 \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ g_5 \\ g_6 \end{bmatrix}$$

where, for $j = 1, ...4$, we have

$$c_{1j} = \frac{\mu_m\Omega^{2+\xi}\,(\ln(\Omega)\,((\nu_m - 1)\,\xi + \nu_m(3\,Q_j - 2)) + (1 - \nu_m)\ln(\omega_j))\,\omega_j}{\mu_i\,\ln(\Omega)\,(2\,\nu_m - 1)} +$$

$$-\frac{\Omega^{1-\xi}\,(\nu_i(9\,Q_j + 1) - 7)\,\omega_j}{10\,\nu_i - 7},$$

$$c_{2j} = \frac{2\,\Omega^{1-\xi}\,((4\,\nu_i - 7)\,Q_j + 6\,\nu_i)\,\omega_j}{10\,\nu_i - 7} - \frac{\mu_m\Omega^{2+\xi}\,(((\xi - 1)\ln(\Omega) - \ln(\omega_j))\,Q_j + 2\ln(\Omega))\,\omega_j}{\mu_i\,\ln(\Omega)},$$

$$c_{3j} = -6\,\nu_m\,(\nu_m - 1)\,Q_j + (\nu_m - 1)\,(\xi + 4 - 4\,\nu_m) + \frac{(1 - \nu_m)\ln(\omega_j)}{\ln(\Omega)},$$

$$c_{35} = 5\,(2\,\nu_m - 1)\,(\nu_m - 1), \quad c_{36} = \frac{10\,(2\,\nu_m - 1)^2\,(\nu_m - 1)}{b^3\Omega^3\,(4\,\nu_m - 5)}$$

$$c_{4j} = (4\,\nu_m + \xi - 3)\,Q_j + \frac{8}{3}\nu_m - \frac{14}{3} - \frac{\ln(\omega_j)}{\ln(\Omega)}Q_j,$$

$$c_{45} = \frac{20}{3}(1 - \nu_m), \quad c_{46} = -\frac{20\,(\nu_m - 1)}{3\,b^3\Omega^3}.$$

Respectively, for the displacement or the traction boundary conditions, the terms of the last two lines are

$$c_{5j}^{(u)} = \frac{6\,\nu_m\left(R^7 - b^7\Omega^7\right)Q_j + b^7\Omega^7\,(7 - 4\,\nu_m) + 4\,R^7\nu_m}{7\,R^5 b^2 \Omega^2},$$

$$c_{55}^{(u)} = 1 - \frac{10\,R^2\nu_m}{7\,b^2\Omega^2} + \frac{(10\,\nu_m - 7)\,\Omega^5 b^5}{7\,R^5},$$

$$c_{56}^{(u)} = \frac{5\,b^2\,(4\,\nu_m\,(2\,\nu_m - 3) + 7)\,\Omega^2}{7\,R^5\,(4\,\nu_m - 5)} + \frac{1}{R^3} - \frac{8\,R^2\nu_m\,(5\,\nu_m - 4)}{7\,b^5\,(4\,\nu_m - 5)\,\Omega^5}, \quad g_5 = g_5^{(u)} = \gamma,$$

$$c_{6j}^{(u)} = \frac{\Omega^5 b^5\,(\nu_m\,(6\,Q_j + 4) - 7)}{R^5\,(10\,\nu_m - 7)}, \quad c_{65}^{(u)} = 1 - \frac{\Omega^5 b^5}{R^5},$$

$$c_{66}^{(u)} = \frac{5\left(R^2 - \Omega^2 b^2\right)(4\,\nu_m\,(2\,\nu_m - 3) + 7)}{R^5\,(4\,\nu_m - 5)\,(10\,\nu_m - 7)}, \quad g_6 = g_6^{(u)} = \gamma,$$

or

$$c_{5j}^{(T)} = -\frac{\nu_m\,(3\,Q_j + 2)\,R^2\mu_m}{7\,\Omega^2 b^2} + \frac{4\,\Omega^5 b^5\,(\nu_m\,(6\,Q_j + 4) - 7)\,\mu_m}{7\,R^5},$$

$$c_{55}^{(T)} = \frac{\left(4\,b^7\,(7 - 10\,\nu_m)\,\Omega^7 + R^5\,\left(7\,\Omega^2 b^2 + 5\,R^2\nu_m\right)\right)\mu_m}{7\,R^5 b^2\Omega^2},$$

$$c_{56}^{(T)} = \frac{-2\mu_m\left(10\left(8\,b^7\Omega^7 - R^7\right)\nu_m{}^2 + \left(\Omega^5 b^5\left(-120\,\Omega^2 b^2 + 7\,R^2\right) + 8\,R^7\right)\nu_m\right)}{7\,R^5\Omega^5 b^5\,(4\,\nu_m - 5)} +$$

$$-\frac{10\,\mu_m\left(2\,\Omega^2 b^2 - R^2\right)}{R^5\,(4\,\nu_m - 5)}, \quad g_5 = g_5^{(T)} = \tau,$$

$$c_{6j}^{(T)} = \frac{4\,\Omega^5 b^5\,(\nu_m\,(6\,Q_j + 4) - 7)}{R^5\,(5\,\nu_m + 7)}, \quad c_{65}^{(T)} = 1 - \frac{4\,(10\,\nu_m - 7)\,\Omega^5 b^5}{R^5\,(5\,\nu_m + 7)},$$

$$c_{66}^{(T)} = \frac{-20\,b^2\,(4\,\nu_m\,(2\,\nu_m - 3) + 7)\,\Omega^2 + 10\,\left(7 - \nu_m{}^2\right)R^2}{R^5\,(4\,\nu_m - 5)\,(5\,\nu_m + 7)}, \quad g_6 = g_6^{(T)} = -\frac{\tau}{\mu_m}.$$

3. Quantities related to the shear bounds

We introduce the following quantities:

$$V_1 = V_{7,1}V_{8,3} - V_{7,3}V_{8,1}, \quad V_2 = V_{7,1}V_{10,3} - V_{7,3}V_{10,1},$$

$$V_3 = V_{8,3}V_{10,1} - V_{8,1}V_{10,3}, \quad V_4 = V_{7,1}V_{9,3} - V_{7,3}V_{9,1},$$

$$V_5 = V_{9,3}V_{8,1} - V_{9,1}V_{8,3}, \quad V_6 = V_{9,1}V_{10,3} - V_{9,3}V_{10,1}$$

and

$$V_{7,1} = s_{14} + s_{24} + 1, \quad V_{7,3} = s_{13} + s_{23} + 1,$$

$$V_{8,1} = Q_1 s_{14} + Q_2 s_{24} + Q_4, \quad V_{8,3} = Q_1 s_{13} + Q_2 s_{23} + Q_3,$$

$$V_{9,1} = -\left(\nu_m - 1\right)\left(\Lambda_1 + \left(s_{14} - s_{24}\right)\Lambda_2\right) + 3\left(Q_1 s_{14} + Q_2 s_{24} + Q_4\right)\nu_m +$$

$$+ \left(\left(\nu_m - 1\right)\xi - 2\nu_m\right)\left(s_{14} + s_{24} + 1\right),$$

$$V_{9,3} = \left(\nu_m - 1\right)\left(\Lambda_1 + \left(s_{13} - s_{23}\right)\Lambda_2\right) + 3\left(Q_1 s_{13} + Q_2 s_{23} + Q_3\right)\nu_m +$$

$$+ \left(\left(\nu_m - 1\right)\xi - 2\nu_m\right)\left(s_{13} + s_{23} + 1\right),$$

$$V_{10,1} = \left(Q_1 s_{14} - Q_2 s_{24}\right)\mu_m \Lambda_2 - \Lambda_1 Q_4 \mu_m +$$

$$+ \mu_m \left(\left(Q_1 s_{14} + Q_2 s_{24} + Q_4\right)\left(\xi - 1\right) - 10/3(s_{14} + s_{24} + 1)\right),$$

$$V_{10,3} = \left(Q_1 s_{13} - Q_2 s_{23}\right)\mu_m \Lambda_2 + Q_3 \mu_m \Lambda_1 +$$

$$+ \mu_m \left(\left(Q_1 s_{13} + Q_2 s_{23} + Q_3\right)\left(\xi - 1\right) - 10/3(s_{13} + s_{23} + 1)\right),$$

where

$$s_{13} = \frac{\alpha_{12}\alpha_{23} - \alpha_{13}\alpha_{22}}{\alpha_{11}\alpha_{22} - \alpha_{12}\alpha_{21}}, \quad s_{14} = \frac{\alpha_{12}\alpha_{24} - \alpha_{14}\alpha_{22}}{\alpha_{11}\alpha_{22} - \alpha_{12}\alpha_{21}},$$

$$s_{23} = \frac{\alpha_{13}\alpha_{21} - \alpha_{11}\alpha_{23}}{\alpha_{11}\alpha_{22} - \alpha_{12}\alpha_{21}}, \quad s_{24} = \frac{\alpha_{14}\alpha_{21} - \alpha_{11}\alpha_{24}}{\alpha_{11}\alpha_{22} - \alpha_{12}\alpha_{21}}$$

and, for $j = 1, ...4$, we have

$$\alpha_{1j} = -\left(3\nu_m Q_j + \left(\nu_m - 1\right)\xi - \frac{\left(\ln\left(\omega_j\right) + 2\ln\left(\Omega\right)\right)\nu_m}{\left(\ln\left(\Omega\right)\right)^2} + \frac{\ln\left(\omega_j\right)}{\ln\left(\Omega\right)}\right)\left(10\nu_i - 7\right)\Omega^{2\xi+1}\mu_m \omega_j +$$

$$+ \left(2\nu_m - 1\right)\left(\nu_i(9Q_j + 1) - 7\right)\mu_i \omega_j,$$

$$\alpha_{2j} = -\left(2 + \left(\xi - 1 - \frac{\ln\left(\omega_j\right)}{\ln\left(\Omega\right)}\right)Q_j\right)\left(10\nu_i - 7\right)\Omega^{2\xi+1}\mu_m \omega_j + 8\left(\left(Q_j + \frac{3}{2}\right)\nu_i - \frac{7}{4}Q_j\right)\mu_i \omega_j.$$

Funding statement

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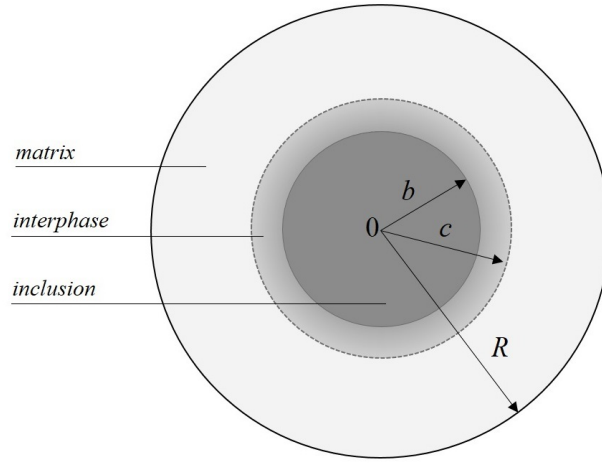
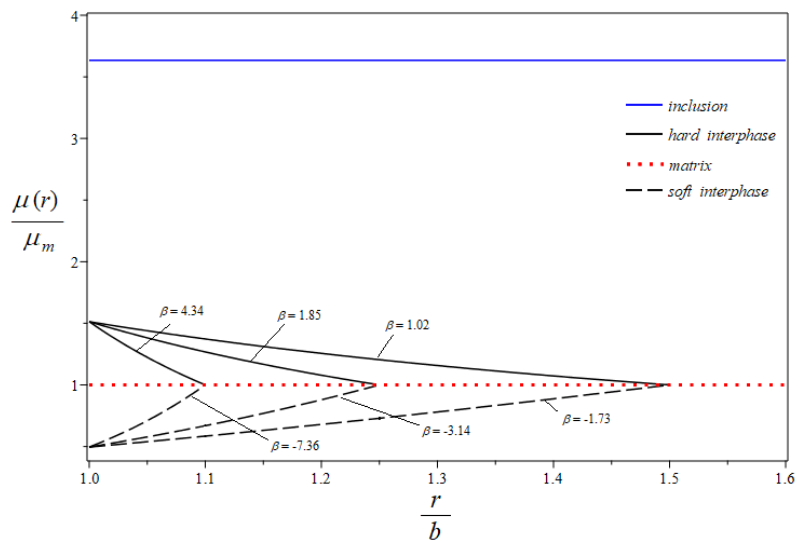Figure 1. Composite sphere model in the plane $z = 0$

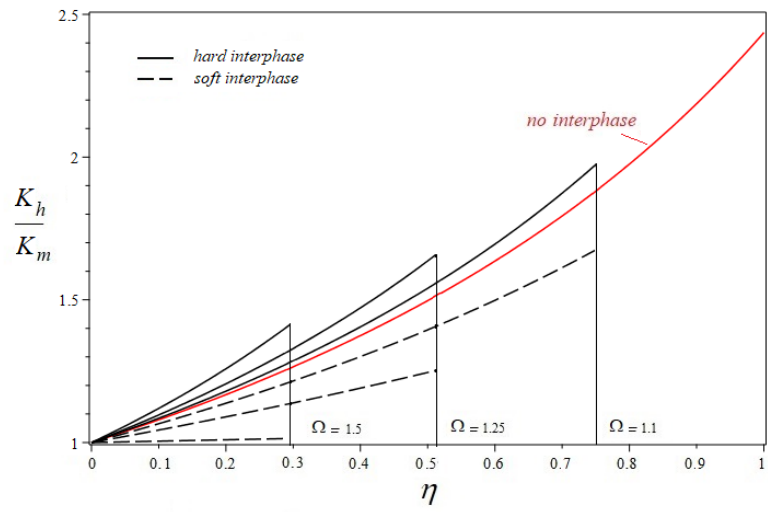Figure 2. Power laws in the graded interphase zone for hard and soft interphases

Figure 3. Normalized bulk modulus for varying interphase thicknesses for hard and soft interphases
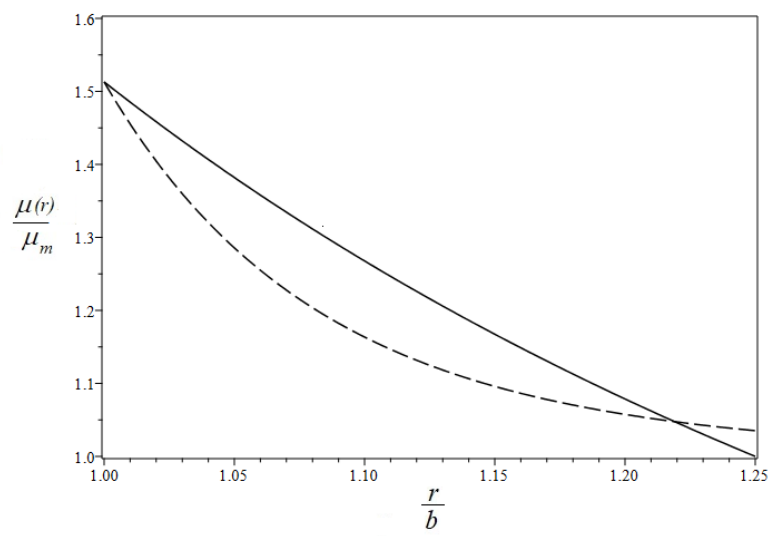
Figure 4. Power laws for the graded interphase ($\Omega = 1.25$) with different elastic modulus property: present model (solid line) and model of Sburlati and Cianci (2015) (dashed line)
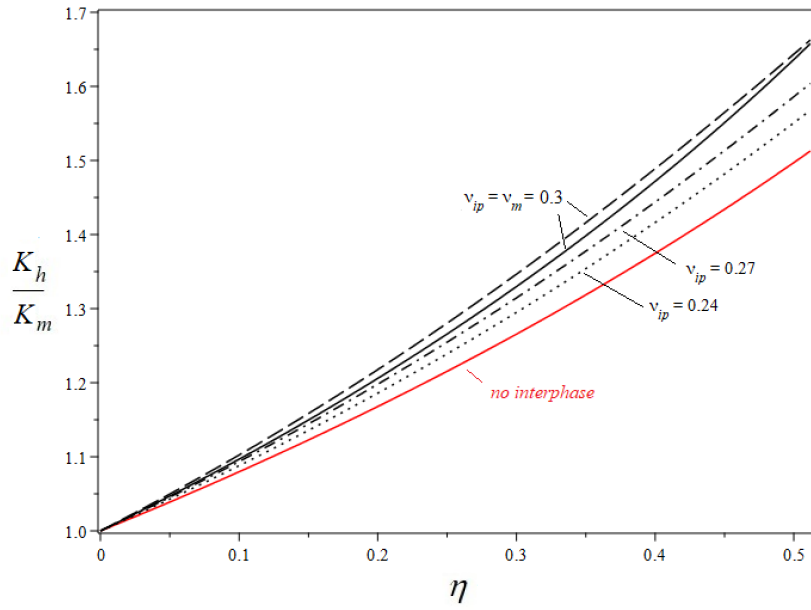
Figure 5. Effects of graded Poisson's ratio on the effective bulk modulus. Comparisons with the results obtained with the present model (solid line) and the results obtained with the Sburlati and Cianci (2015) model assuming: $\nu_{ip} = \nu_m = 0.3$ (dashed line), $\nu_{ip} = 0.27, \nu_m = 0.3$ (dot-dashed line) and $\nu_{ip} = 0.24, \nu_m = 0.3$ (dotted line)

Figure 6. Normalized shear modulus bounds for varying interphase thicknesses with hard (a) or soft interphase (b)

Figure 1: *Composite sphere model in the plane $z = 0$*

Figure 2: *Power laws in the graded interphase zone for hard and soft interphases*

Figure 3: *Normalized bulk modulus for varying interphase thicknesses for hard and soft interphases*

Figure 4: *Power laws for the graded interphase* ($\Omega = 1.25$) *with different elastic modulus property: present model (solid line) and model of Sburlati and Cianci (2015) (dashed line)*

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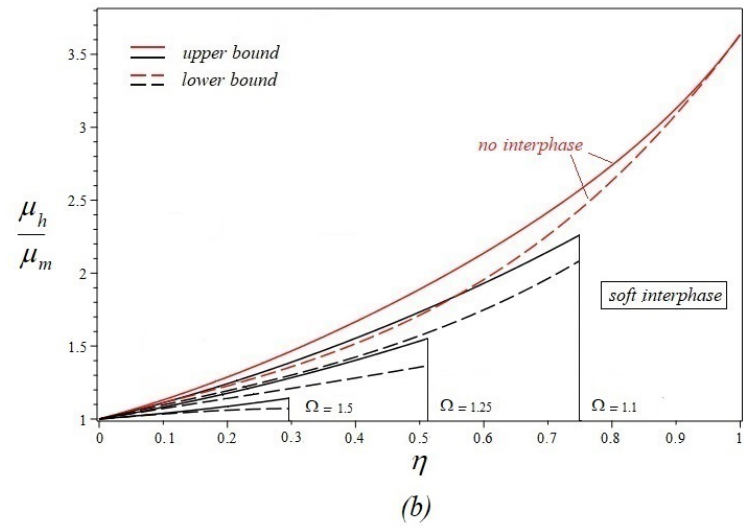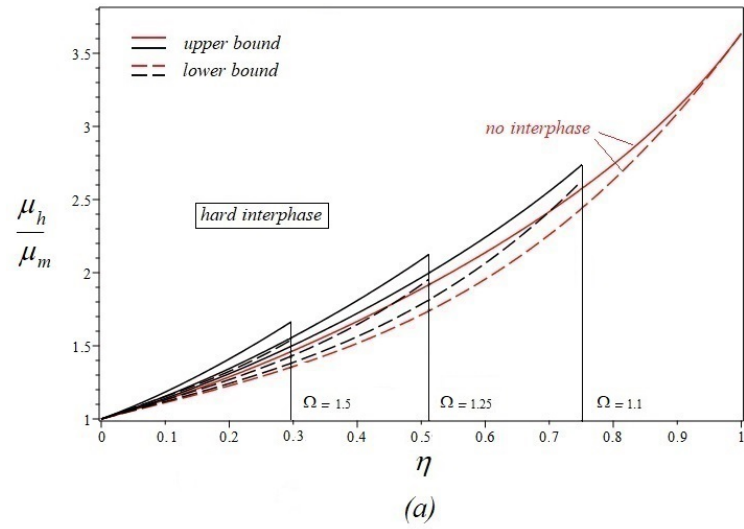
Figure 6: *Normalized shear modulus bounds for varying interphase thicknesses with hard (a) or soft interphase (b)*