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European Journal of Mechanics A/Solids 22 (2003) 885–902

EUROPEAN JOURNAL OF MECHANICS A/SOLIDS

Differential schemes for the elastic characterisation of dispersions of randomly oriented ellipsoids

Stefano Giordano

Department of Biophysical and Electronic Engineering, University of Genoa, Via Opera Pia 11 A, 16145 Genoa, Italy

Received 19 May 2003; accepted 25 July 2003

Abstract

The paper deals with the elastic characterisation of dispersions of randomly oriented ellipsoids: we start from the theory of strongly diluted mixtures and successively we generalise it with a differential scheme. The micro-mechanical averaging inside the composite material is carried out by means of explicit results which allows us to obtain closed-form expressions for the macroscopic or equivalent elastic moduli of the overall composite materials. This micromechanical technique has been explicitely developed for describing embeddings of randomly oriented not spherical objects. In particular, this study has been applied to characterise media with different shapes of the inclusions (spheres, cylinders and planar inhomogeneities) and for special media involved in the mixture definition (voids or rigid particles): an accurate analysis of all these cases has been studied yielding a set of relations describing several composite materials of great technological interest. The differential effective medium scheme (developed for generally shaped ellipsoids) extends such results to higher values of the volume fraction of the inhomogeneities embedded in the mixture. For instance, the analytical study of the differential scheme for porous materials (with ellipsoidal zero stiffness voids) reveals a universal behaviour of the effective Poisson ratio for high values of the porosity. This means that Poisson ratio at high porosity assumes characteristic values depending only on the shape of the inclusions and not on the elastic response of the matrix.

Keywords: Composite materials; Elastic homogenisation and mixture theory

1. Introduction

In recent years the characterisation of heterogeneous materials has attracted an ever increasing interest. A central problem, of considerable technological importance, is to evaluate the effective elastic properties governing the behaviour of a composite material on the macroscopic scale. At present, it is well known that it does not exist a universally applicable mixing formula giving the effective properties of the heterogeneous materials as some sort of average of the properties of the constituent materials. Actually, the details of the micro-geometry can play a crucial role in determining the overall properties. Therefore, the elastic (thermal, electrical and so on) properties of composite materials are strongly microstructure dependent. The relationships between microstructure and properties may be used for designing and improving materials, or conversely, for interpreting experimental data in terms of micro-structural features. A great number of theoretical formulas have been proposed to describe the behaviour of composite materials. A disadvantage of some approximated results is that they do not correspond to *a priori* known microstructure; this kind of results may be interpreted and classified only by means of comparison with numerical or experimental data. A different class of theories is rigorously based on realistic microstructures. These are the classical Hashin and Shtrikman (1962, 1963) variational bounds, which provide an upper and lower bound for composite materials, and the expansions of Brown (1965) and Torquato (1997, 1998) which take into account the spatial correlation function of the

E-mail address: giorda@dibe.unige.it (S. Giordano).

0997-7538/$ – see front matter
doi:10.1016/S0997-7538(03)00091-3

phases. Effective medium theories are commonly used because of their relative simplicity compared to numerical computations: typically, they allow a simple determination of the equivalent elastic properties of a composite material. Therefore, it is very important to establish the conditions of validity and the microstructures for which the theories yield accurate predictions: comparisons with numerical results are necessary and indispensable. So, some efficient numerical algorithms have been produced by Garboczy and Day (1995) and Garboczy and Berryman (2001) to compute the effective linear elastic properties of heterogeneous materials.

Dispersions or suspensions of ellipsoidal inclusions in a homogeneous matrix give a particular example of heterogeneous materials: these media have been extensively studied both from the electrical and the elastic point of view. We briefly describe the earlier literature for electrical and elastic characterisation to draw a comparison between the approaches. From the historical point of view, one of the first attempts to characterise electrical dispersions of spheres is that of Maxwell (1881), which has find out a famous formula for a strongly diluted suspension.

A better model has been provided by the differential scheme, which derives from the mixture characterisation approach used by Bruggeman (1935) and extensively described by Van Beek (1967). In this case the relations should maintain the validity also for less diluted suspensions of spheres. To understand the effects of different shape of the inclusions, ellipsoidal shaped particles have been considered: the first papers dealing with mixtures of ellipsoids were written by Fricke (1924, 1953) concerning the electrical characterisation of biological tissues containing spheroidal particles. In recent literature (Sen et al., 1981; Mendelson and Cohen, 1982; Sen, 1984) several applications of the Bruggeman differential procedure to mixtures of ellipsoids of rotation have been performed in connection with the problem of characterising the dielectric response of water-saturated rocks. A complete differential theory for generally shaped dielectric ellipsoidal inclusions has been developed by Giordano (2003).

Dealing with elastic characterisation of dispersions (see Walpole, 1981; Hashin, 1983) some similar works have been developed: the most famous and studied elastic mixture theory regards a composite material formed by spherical inclusions embedded in a solid matrix. This result is attributed to numerous authors (see Hashin, 1983; Douglas and Garboczi, 1995). To adapt the dilute formulas to the case of any finite volume fraction a great number of proposals have been made and they appear in technical literature. The differential approach is also used in micro-mechanical theories (McLaughlin, 1977; Norris, 1985; Avellaneda, 1987): this leads, in the simpler and most studied case (dispersions of spheres), to a pair of coupled differential equations (see McLaughlin, 1977) which may be numerically solved and the results generate the so-called differential effective medium theory.

Drawing a comparison between the literature dealing with the electrical and the elastic mixture characterisation we may observe that the case of dispersions of ellipsoidal inclusions has not been completely treated from the elastic point of view and the relative differential effective medium scheme has not been developed. Therefore, we devote this paper to fill the gap in this topic. In particular we try to characterise a dispersion of randomly oriented elastic ellipsoids embedded in a given solid matrix. We will obtain a theory for very diluted dispersions similar to the electric Maxwell–Garnett–Fricke theory (see Maxwell and Garnett, 1904; Sihvola, 1999) and we will apply the differential procedure to generalise it to higher values of the volume fraction of the inclusions. To do this, we generalise the well known micromechanical averaging techniques to obtain a specific procedure developed *ad hoc* for embeddings of randomly oriented objects, which is the main purpose of the paper.

2. Theory for strongly diluted dispersions of generally shaped ellipsoids

The elastic properties of two-phase materials depend on the geometrical nature of the mixture (microstructure) and on the volume fraction of the two media. Such a composite material can be thought as a heterogeneous solid continuum that bonds together two homogeneous continua: each part of the media has a well-defined sharp boundary. The bonding at the interfaces remains intact in our models when the whole mixture is placed in an equilibrated state of infinitesimal elastic strain by external loads or constraints. In the present case, the boundary conditions require that both the vector displacement and the stress tensor be continuous across any interfaces. Each separate homogeneous region is characterised by its stiffness tensor, which describes the stress-strain relation. If both materials are linear and homogeneous this relation is given by:

$$T_{ij} = L^s_{ijkl} E_{kl}, \quad s = 1, 2, \tag{1}$$

where $\mathbf{T}$ is the stress tensor (3×3 sized), $\mathbf{E}$ is the strain tensor (3×3 sized) and $\mathbf{L}$ is the constant stiffness tensor, which depends on the medium considered ($s = 1, 2$). For isotropic media this latter is written, for example in terms of the bulk and shear constants, as follows:

$$L^s_{ijkl} = k_s \delta_{ij} \delta_{kl} + 2\mu_s \left(\delta_{ik} \delta_{jl} - \frac{1}{3} \delta_{ij} \delta_{kl} \right), \quad s = 1, 2, \tag{2}$$

where k_s and μ_s are the bulk and shear moduli of the s-th medium ($s = 1, 2$) and δ_{nm} is the Kronecker's delta. To solve a mixture problem consists in finding the equivalent macroscopic stiffness tensor for the whole composite material and then, for overall isotropic behaviour, this means that we have to evaluate the equivalent k and μ constants. Here we consider a dispersion of randomly oriented ellipsoids and firstly we develop a procedure to evaluate the average value of the strain inside each inclusion embedded in a given matrix with a bulk strain applied to the overall structure. We start with some definitions used to simplify the problem. Instead of describing the strain with the complete symmetric tensor we adopt a column vector, which contains the six independent elements in a given order; the same approach is used for the stress (T means transposed):

$$\widehat{\mathbf{E}} = [\,E_{11} \quad E_{22} \quad E_{33} \quad E_{12} \quad E_{23} \quad E_{13}\,]^{\mathrm{T}}; \qquad \widehat{\mathbf{T}} = [\,T_{11} \quad T_{22} \quad T_{33} \quad T_{12} \quad T_{23} \quad T_{13}\,]^{\mathrm{T}}. \tag{3}$$

Adopting this notation scheme the stiffness four-index tensor for the isotropic components is represented by a simpler matrix with six rows and six columns:

$$\hat{\mathbf{L}}^s = \begin{bmatrix} k_s + \frac{4}{3}\mu_s & k_s - \frac{2}{3}\mu_s & k_s - \frac{2}{3}\mu_s & 0 & 0 & 0 \\ k_s - \frac{2}{3}\mu_s & k_s + \frac{4}{3}\mu_s & k_s - \frac{2}{3}\mu_s & 0 & 0 & 0 \\ k_s - \frac{2}{3}\mu_s & k_s - \frac{2}{3}\mu_s & k_s + \frac{4}{3}\mu_s & 0 & 0 & 0 \\ 0 & 0 & 0 & 2\mu_s & 0 & 0 \\ 0 & 0 & 0 & 0 & 2\mu_s & 0 \\ 0 & 0 & 0 & 0 & 0 & 2\mu_s \end{bmatrix}, \quad s = 1, 2, \tag{4}$$

so that the stress-strain relations became $\widehat{\mathbf{T}} = \hat{\mathbf{L}}^1\widehat{\mathbf{E}}$ in the matrix and $\widehat{\mathbf{T}} = \hat{\mathbf{L}}^2\widehat{\mathbf{E}}$ inside each inclusion. To begin the strain computation, we take into consideration a single ellipsoidal isotropic inclusion (medium 2) added to an isotropic matrix (medium 1) placed in an equilibrated state of infinitesimal constant elastic strain. In particular we consider an ellipsoid with axes $a_1,\ a_2,\ a_3$ aligned with the axes $x_1 = x,\ x_2 = y,\ x_3 = z$ of the reference frame with the assumption $a_1 > a_2 > a_3 > 0$ and we define two eccentricities, which describe the shape of the inclusion: $0 < e = a_3/a_2 < 1$ and $0 < g = a_2/a_1 < 1$. It is important to notice that the internal strain is constant if the external or bulk strain is constant. Accordingly with the Eshelby (1957, 1959) theory (extensively described in Mura, 1987; Nemat-Nasser, 1993) the relationship between the uniform original external strain and the induced internal strain is given by:

$$\widehat{\mathbf{E}}_i = \left\{\mathbf{I} - \widehat{\mathbf{S}}\left[\mathbf{I} - \left(\hat{\mathbf{L}}^1\right)^{-1}\hat{\mathbf{L}}^2\right]\right\}^{-1}\widehat{\mathbf{E}}_0 = \hat{\mathbf{A}}\widehat{\mathbf{E}}_0, \tag{5}$$

here $\mathbf{I}$ is the identity matrix with size 6×6, $\widehat{\mathbf{E}}_i$ is the internal strain, $\widehat{\mathbf{E}}_0$ is the original external strain, $\hat{\mathbf{L}}^1$ and $\hat{\mathbf{L}}^2$ are the stiffness tensor of the matrix and the inclusion respectively and $\widehat{\mathbf{S}}$ is the Eshelby tensor, which depends on the eccentricities e and g of the ellipsoid and on the Poisson ratio $\nu_1 = (3k_1 - 2\mu_1)/[2(3k_1 + \mu_1)]$ of the matrix (see Mura, 1987, for a complete description of all the entries and for special cases). Here, we only remember that the general structure of $\widehat{\mathbf{S}}$ is given by:

$$\widehat{\mathbf{S}} = \begin{bmatrix} s_{1111} & s_{1122} & s_{1133} & 0 & 0 & 0 \\ s_{2211} & s_{2222} & s_{2233} & 0 & 0 & 0 \\ s_{3311} & s_{3322} & s_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & 2s_{1212} & 0 & 0 \\ 0 & 0 & 0 & 0 & 2s_{2323} & 0 \\ 0 & 0 & 0 & 0 & 0 & 2s_{1313} \end{bmatrix}. \tag{6}$$

Matrix $\hat{\mathbf{A}}$ is simply defined by Eq. (5). We remember that Eq. (5) is written taking into account a particular reference frame with axes aligned to the three principal directions of the embedded ellipsoid. In these condition matrix $\hat{\mathbf{A}}$ has the following mathematical form:

$$\hat{\mathbf{A}} = \begin{bmatrix} A_{1111} & A_{1122} & A_{1133} & 0 & 0 & 0 \\ A_{2211} & A_{2222} & A_{2233} & 0 & 0 & 0 \\ A_{3311} & A_{3322} & A_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & A_{1212} & 0 & 0 \\ 0 & 0 & 0 & 0 & A_{2323} & 0 \\ 0 & 0 & 0 & 0 & 0 & A_{1313} \end{bmatrix}. \tag{7}$$

All the coefficients A_{ijkl} that not appear in Eq. (7) are always zero. With the aim of analysing the behaviour of a mixture of randomly oriented ellipsoids, we need to evaluate the average value of the internal strain inside the ellipsoid over all its possible orientations or rotations in the space. To perform this averaging over all the rotations we name the original reference frame with the letter B and we consider another generic reference frame that is named with the letter F.

The relation between these bases B and F is described by means of a generic rotation matrix $\mathbf{R}(\psi,\theta,\varphi)$ where ψ, θ and φ are the Euler angles; we may consider this matrix as the product of three elementary rotations along the axis z, x and z respectively:

$$\mathbf{R}(\psi,\theta,\varphi)=\begin{bmatrix}\cos\psi & -\operatorname{sen}\psi & 0\\ \operatorname{sen}\psi & \cos\psi & 0\\ 0 & 0 & 1\end{bmatrix}\begin{bmatrix}1 & 0 & 0\\ 0 & \cos\theta & -\operatorname{sen}\theta\\ 0 & \operatorname{sen}\theta & \cos\theta\end{bmatrix}\begin{bmatrix}\cos\varphi & -\operatorname{sen}\varphi & 0\\ \operatorname{sen}\varphi & \cos\varphi & 0\\ 0 & 0 & 1\end{bmatrix}. \tag{8}$$

Therefore the following relations hold on between the different frames: $\mathbf{E}_i^B=\mathbf{R}\mathbf{E}_i^F\mathbf{R}^{\mathrm{T}}$ for the internal strain and $\mathbf{E}_0^B=\mathbf{R}\mathbf{E}_0^F\mathbf{R}^{\mathrm{T}}$ for the bulk strain (here the subscript T means transposed). These expressions have been written with standard notation for the strain (3×3 sized matrix). They may be converted in our notation defining a matrix $\widehat{\mathbf{M}}(\psi,\theta,\varphi)$, 6×6 sized, which acts as a rotation matrix on our strain vectors: so, we may write $\widehat{\mathbf{E}}_i^B=\widehat{\mathbf{M}}\widehat{\mathbf{E}}_i^F$ inside the ellipsoid and $\widehat{\mathbf{E}}_0^B=\widehat{\mathbf{M}}\widehat{\mathbf{E}}_0^F$ outside it. The entries of the matrix $\widehat{\mathbf{M}}$ are completely defined by the comparison between the relations $\mathbf{E}_i^B=\mathbf{R}\mathbf{E}_i^F\mathbf{R}^{\mathrm{T}}$ and $\widehat{\mathbf{E}}_i^B=\widehat{\mathbf{M}}\widehat{\mathbf{E}}_i^F$ and by considering the notation adopted for the strain. Eq. (5) is written on the frame B and therefore it actually reads $\widehat{\mathbf{E}}_i^B=\hat{\mathbf{A}}\widehat{\mathbf{E}}_0^B$; this latter may be reformulated on the generic frame F simply obtaining:

$$\widehat{\mathbf{E}}_i^F=\left\{\widehat{\mathbf{M}}(\Psi,\theta,\varphi)^{-1}\hat{\mathbf{A}}\widehat{\mathbf{M}}(\Psi,\theta,\varphi)\right\}\widehat{\mathbf{E}}_0^F. \tag{9}$$

Finally, the average value of the strain inside the inclusion may be computed by means of the following integration over all the possible rotations:

$$\langle\widehat{\mathbf{E}}_i\rangle=\frac{1}{8\pi^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}\left\{\widehat{\mathbf{M}}(\Psi,\theta,\varphi)^{-1}\hat{\mathbf{A}}\widehat{\mathbf{M}}(\Psi,\theta,\varphi)\right\}\operatorname{sen}\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi\,\mathrm{d}\Psi\,\widehat{\mathbf{E}}_0. \tag{10}$$

By means of a very long but straightforward integration we have obtained an explicit relation between the external strain $\widehat{\mathbf{E}}_0$ $(=\widehat{\mathbf{E}}_0^F)$ and the average value $\langle\widehat{\mathbf{E}}_i\rangle$ inside the randomly oriented ellipsoid:

$$\langle\widehat{\mathbf{E}}_i\rangle=\begin{bmatrix}\alpha & \beta & \beta & 0 & 0 & 0\\ \beta & \alpha & \beta & 0 & 0 & 0\\ \beta & \beta & \alpha & 0 & 0 & 0\\ 0 & 0 & 0 & \alpha-\beta & 0 & 0\\ 0 & 0 & 0 & 0 & \alpha-\beta & 0\\ 0 & 0 & 0 & 0 & 0 & \alpha-\beta\end{bmatrix}\widehat{\mathbf{E}}_0=\widehat{\mathbf{B}}\widehat{\mathbf{E}}_0, \tag{11}$$

where α and β depend only on the coefficients A_{ijkl}:

$$\begin{cases}\alpha = \frac{1}{5}A_{1111}+\frac{1}{5}A_{2222}+\frac{1}{5}A_{3333}+\frac{2}{15}A_{1313}+\frac{2}{15}A_{2323}+\frac{2}{15}A_{1313}\\ \qquad +\frac{1}{15}A_{1122}+\frac{1}{15}A_{1133}+\frac{1}{15}A_{2211}+\frac{1}{15}A_{2233}+\frac{1}{15}A_{3311}+\frac{1}{15}A_{3322},\\ \beta = \frac{1}{15}A_{1111}+\frac{1}{15}A_{2222}+\frac{1}{15}A_{3333}-\frac{1}{15}A_{1313}-\frac{1}{15}A_{2323}-\frac{1}{15}A_{1313}\\ \qquad +\frac{2}{15}A_{1122}+\frac{2}{15}A_{1133}+\frac{2}{15}A_{2211}+\frac{2}{15}A_{2233}+\frac{2}{15}A_{3311}+\frac{2}{15}A_{3322}.\end{cases} \tag{12}$$

This is the main result of this section and it plays a crucial role in the further development of the theory. To sum up, we evaluate the matrix $\hat{\mathbf{A}}$ by means of Eq. (5) and then we apply Eq. (12) to average the internal strain over all the possible orientation of the ellipsoid. We wish to point out that Eq. (12) is extremely convenient to perform the micro mechanical averaging because it removes the problem of the integral evaluation and allows us to obtain results in closed form. Of course, standard micro mechanical techniques are largely discussed and applied in earlier literature (Mura, 1987; Nemat-Nasser, 1993); however, the averaging of the strain over the geometric orientations of the randomly oriented microinclusions, explicitly performed by means of Eq. (12), is an useful result which permits to work out in detail several analysis of various kind of composite materials: the explicit result of general validity may be used to avoid the complicated integration over the angles which define the orientation of the elastic microinclusions. More precisely, the sums appearing for example in Eqs. (7.4.6a,b) or (7.4.9a,b) (equation numbering in the book by Nemat-Nasser, 1993) become integrals for randomly oriented inclusions and the proposed approach solves the problem of their calculations. With Eq. (12) the sole knowledge of the Eshelby tensor of the inclusion permits, with algebraic computations, to characterise the overall dilute dispersion of randomly oriented generally shaped ellipsoids. Moreover, the

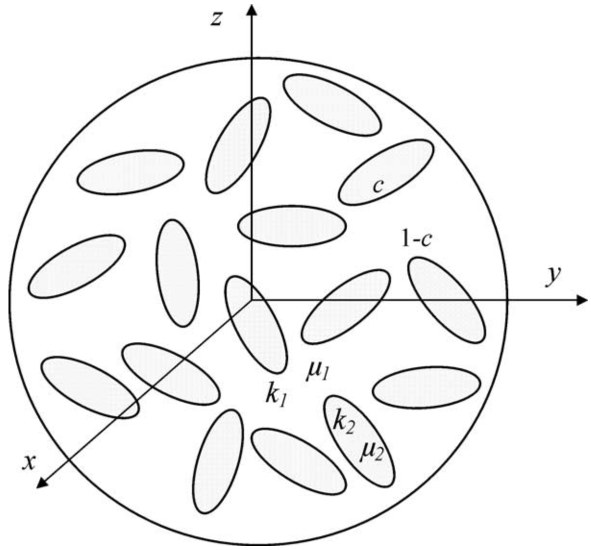


Fig. 1. Structure of a dispersion of randomly oriented ellipsoids. The external surface of the mixture is, e.g., a sphere, which contains all the inclusions. The two-phase material is described by the elastic response of each phase, by the volume fraction of the inclusions and by the characteristic eccentricities of the embedded particles.

definition of the coefficients α and β by means of Eq. (12) is useful to introduce the differential scheme for randomly oriented generally shaped ellipsoids in a very simple way (see next sections).

Still now, we have considered a single ellipsoidal particle and we have calculated the averaged internal strain $\langle\widehat{\mathbf{E}}_i\rangle$ when it is randomly placed in a matrix with a given constant strain $\widehat{\mathbf{E}}_0$; from now on, we have to deal with an ensemble of inclusions randomly oriented and distributed in the solid matrix (see Fig. 1). To begin the analysis of the behaviour of the overall structure, first of all, we consider an extremely low value of the volume fraction of the dispersed component so that we may neglect the interactions among the inclusions. Therefore, each ellipsoidal particle behaves as a single one in the whole space. We define c as the volume fraction of the inclusions. Because of the low value of c we may compute the average value of the elastic strain over the whole heterogeneous material by means of the relation:

$$\langle\widehat{\mathbf{E}}\rangle = (1-c)\widehat{\mathbf{E}}_0 + c\langle\widehat{\mathbf{E}}_i\rangle = \left[(1-c)\mathbf{I} + c\widehat{\mathbf{B}}\right]\widehat{\mathbf{E}}_0, \tag{13}$$

where we have considered the strain outside the inclusions approximately constant and identical to the bulk strain $\widehat{\mathbf{E}}_0$ (matrix $\widehat{\mathbf{B}}$ is defined by Eq. (11)). Moreover, we define $\hat{\mathbf{L}}_{eq}$ as the equivalent stiffness tensor of the whole mixture (which is isotropic because of the randomness of the orientations of the inclusions) by means of the relation $\langle\widehat{\mathbf{T}}\rangle = \hat{\mathbf{L}}_{eq}\langle\widehat{\mathbf{E}}\rangle$; to evaluate $\hat{\mathbf{L}}_{eq}$ we need the average value $\langle\widehat{\mathbf{T}}\rangle$ of the stress inside the random material. So, we define V as the total volume of the mixture, V_e as the total volume of the embedded ellipsoids and V_0 as the volume of the remaining space among the inclusions ($V = V_e \cup V_0$). The average value of $\widehat{\mathbf{T}} = \hat{\mathbf{L}}(\bar{r})\widehat{\mathbf{E}}$ over the volume of the whole material is evaluated as follows ($\hat{\mathbf{L}}(\bar{r}) = \hat{\mathbf{L}}^1$ if $\bar{r} \in V_0$ and $\hat{\mathbf{L}}(\bar{r}) = \hat{\mathbf{L}}^2$ if $\bar{r} \in V_e$):

$$\begin{aligned}
\langle\widehat{\mathbf{T}}\rangle &= \frac{1}{V}\int_V \hat{\mathbf{L}}(\bar{r})\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} = \frac{1}{V}\hat{\mathbf{L}}^1\int_{V_0}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\hat{\mathbf{L}}^2\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} \\
&= \frac{1}{V}\hat{\mathbf{L}}^1\int_{V_0}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\hat{\mathbf{L}}^2\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\hat{\mathbf{L}}^1\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} - \frac{1}{V}\hat{\mathbf{L}}^1\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} \\
&= \hat{\mathbf{L}}^1\langle\widehat{\mathbf{E}}\rangle + c\left(\hat{\mathbf{L}}^2 - \hat{\mathbf{L}}^1\right)\langle\widehat{\mathbf{E}}_i\rangle = \hat{\mathbf{L}}^1\langle\widehat{\mathbf{E}}\rangle + c\left(\hat{\mathbf{L}}^2 - \hat{\mathbf{L}}^1\right)\widehat{\mathbf{B}}\widehat{\mathbf{E}}_0.
\end{aligned} \tag{14}$$

Drawing a comparison between Eqs. (13) and (14) we may find a complete expression, which allows us to estimate the equivalent stiffness tensor $\hat{\mathbf{L}}_{eq}$:

$$\hat{\mathbf{L}}_{eq} = \hat{\mathbf{L}}^1 + c\left(\hat{\mathbf{L}}^2 - \hat{\mathbf{L}}^1\right)\widehat{\mathbf{B}}\left[(1-c)\mathbf{I} + c\widehat{\mathbf{B}}\right]^{-1}. \tag{15}$$

Eq. (15) may be written in explicit form finding out all the components of the stiffness tensor; with straightforward algebraic computation, we observe that $\hat{\mathbf{L}}_{\mathrm{eq}}$ corresponds to an isotropic material described by the following bulk and shear moduli:

$$
\begin{aligned}
k_{\mathrm{eq}} &= \frac{k_1(1-c)+ck_2(\alpha+2\beta)}{1-c+c(\alpha+2\beta)} = k_1 + (\alpha+2\beta)(k_2-k_1)c + \mathrm{O}\big(c^2\big),\\
\mu_{\mathrm{eq}} &= \frac{\mu_1(1-c)+c\mu_2(\alpha-\beta)}{1-c+c(\alpha-\beta)} = \mu_1 + (\alpha-\beta)(\mu_2-\mu_1)c + \mathrm{O}\big(c^2\big).
\end{aligned}
\tag{16}
$$

These equations complete the characterisation of a strongly diluted dispersion of randomly oriented ellipsoids: to sum up the theoretical procedure, we remember that the first step consists in evaluating the matrix $\hat{\mathbf{A}}$ defined by Eq. (5), then we use Eq. (12) to compute the values of the coefficients α and β and finally we obtain the equivalent moduli of the overall isotropic structure by means of Eq. (16). Moreover, we observe that the coefficients α and β depend on $k_1, k_2, \mu_1, \mu_2, e, g$ and therefore the characterisation depends on the shape of the inclusions, i.e., on the microscopic morphology of the heterogeneous material. Finally, k_{eq} and μ_{eq} have been expressed in terms of $k_1, k_2, \mu_1, \mu_2, e, g$ and the volume fraction c. We want to point out that results stated in Eq. (16) are the elastic counterpart of the Maxwell–Garnett–Fricke relations (Sihvola, 1999) for the electrical characterisation of ellipsoidal dispersions. Furthermore, it seems

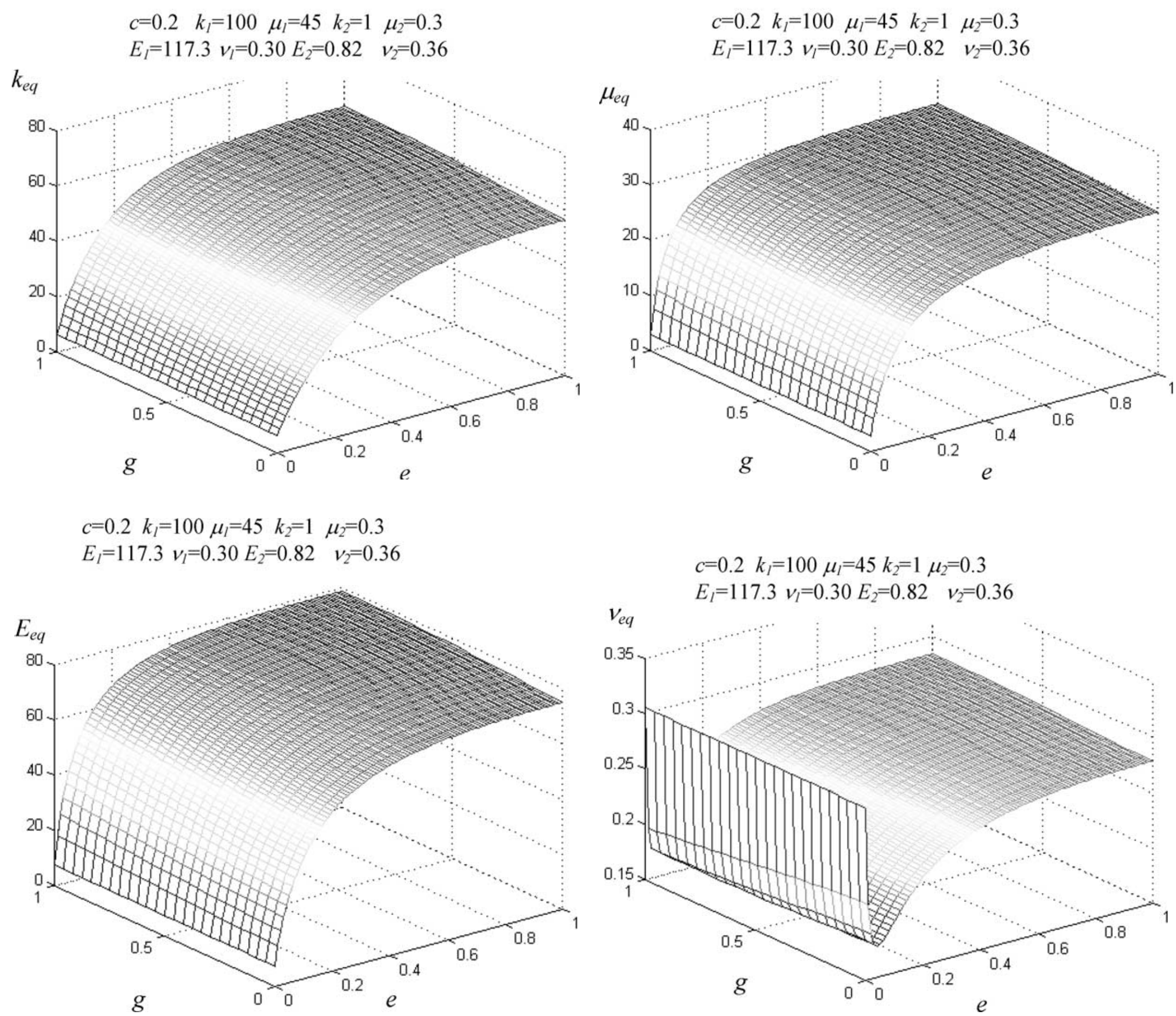


Fig. 2. Effective elastic response for a given dispersion versus the eccentricities e and g of the embedded inclusions (ellipsoids with axes a_1, a_2, a_3 with $a_1 > a_2 > a_3 > 0$. Eccentricities: $0 < e = a_3/a_2 < 1$ and $0 < g = a_2/a_1 < 1$). Bulk, shear, Young and Poisson constants have been computed with the theory for diluted dispersions (Eq. (16)) with the indicated assumptions.

interesting to underline the following fact: dealing with a dielectric ellipsoid we may compute the average value of the electrical field inside the randomly oriented inclusion considering the mean value of the three cases obtained when the ellipsoid is aligned with the three axes of the frame as explained in Fricke (1953) or in Giordano (2003); in our elastic theory this is not true and the correct average value of the strain must be computed with Eq. (12) which is more complicated. This depends on the intrinsic tensor character of the quantities involved in the elastic theory, which not permits to apply simple intuitive or empirical consideration in averaging stress or strain inside a given material. For example, if we use the mean value over the elementary three position of ellipsoids aligned along the three axes, we are actually considering a special dispersion in which the ellipsoids may assume only these three particular positions and therefore, we obtain, as result, a cubic crystal described by three independent parameters instead of the two characteristic moduli of an isotropic medium.

Adopting the general structure of the Eshelby tensor (see Mura, 1987; Nemat-Nasser, 1993) we may theoretically or numerically apply the outlined procedure to evaluate the effects of the shape of the inclusions on the overall elastic behaviour of the composite material. In Fig. 2 one can find some plots that show the behaviour of the elastic moduli versus the eccentricities e and g of the embedded ellipsoids (we consider an ellipsoid with axes $a_1,\ a_2,\ a_3,\ a_1 > a_2 > a_3 > 0$ and we define two eccentricities: $0 < e = a_3/a_2 < 1$ and $0 < g = a_2/a_1 < 1$). In particular we may observe the effect of the shape of the inclusions on the bulk and shear moduli or on the Young modulus and Poisson ratio. It seems interesting to note that the equivalent Poisson ratio, in general, has not a monotonic behaviour with respect to the eccentricities and it may assume values out of the range between ν_1 and ν_2 (Poisson ratio of the matrix and of the inhomogeneities respectively). Other examples will be explained in further sections. We want to point out that these results, still now, are valid only for strongly diluted dispersions; in the next section we present a study of several limiting cases based on this theory (with very low values of the volume fraction) and in the sequel we will describe a more refined differential scheme which should hold on for any concentration of the dispersed phase.

3. Limiting cases: dispersions of inclusions with special shapes

By introducing particular values of the characteristic eccentricities we may find out explicit relations for dispersions of spheres ($e = 1,\ g = 1$), randomly oriented circular cylinder ($e = 1,\ g = 0$) and randomly oriented planar inclusions (slabs or sheets with $e = 0,\ g = 1$). It is important to observe that all the planar structure with $e = 0$ behave in the same way independently on the value of g. Here, it is convenient to think that the value $e = 0$ (for any g) corresponds to infinite two-dimensional planes (sheets) of medium 2 randomly embedded in the matrix (medium 1). This is coherent with some explicit results described in the sequel.

Dealing with suspensions of spheres, the theoretical procedure outlined in the previous section may be explicitly performed by introducing the pertinent form of the Eshelby tensor reported in Mura (1987). Actually, for spheres the averaging technique over all the orientations is not necessary but it may be used as check of the procedure. Anyway, the final result is given by the following relationships:

$$\begin{aligned} k_{\mathrm{eq}} &= \frac{k_1(4\mu_1+3k_2)+4c\mu_1(k_2-k_1)}{4\mu_1+3k_2-3c(k_2-k_1)} = k_1 + \frac{4\mu_1+3k_1}{4\mu_1+3k_2}(k_2-k_1)c + \mathrm{O}(c^2), \\ \mu_{\mathrm{eq}} &= \mu_1 \frac{[(1-c)\mu_1+c\mu_2](9k_1+8\mu_1)+6\mu_2(k_1+2\mu_1)}{\mu_1(9k_1+8\mu_1)+6[(1-c)\mu_2+c\mu_1](k_1+2\mu_1)} \\ &= \mu_1 + \frac{5\mu_1(4\mu_1+3k_1)(\mu_2-\mu_1)}{\mu_1(9k_1+8\mu_1)+6\mu_2(k_1+2\mu_1)}c + \mathrm{O}(c^2). \end{aligned} \tag{17}$$

This is the well known result obtained by several authors in the earlier literature as described in Hashin (1983) and in Douglas and Garboczi (1995). If we consider the characteristic Eshelby tensor for cylinders and we apply the previously outlined procedure, in closed form, we obtain, after a long and tedious algebraic computation, the explicit results:

$$\begin{aligned} k_{\mathrm{eq}} &= k_1 + \frac{\mu_2+3\mu_1+3k_1}{\mu_2+3\mu_1+3k_2}(k_2-k_1)c + \mathrm{O}(c^2), \\ \mu_{\mathrm{eq}} &= \mu_1 + \tfrac{1}{5}(\mu_2-\mu_1)\big\{64\mu_1^4 + 84k_1\mu_1^3 + 63\mu_1^3k_2 + 184\mu_1^3\mu_2 + 156\mu_1^2k_2\mu_2 \\ &\quad + 72\mu_1^2\mu_2^2 + 120k_1\mu_1^2\mu_2 + 81k_1\mu_1^2k_2 + 36k_1\mu_2^2\mu_1 + 90k_1k_2\mu_1\mu_2 + 21k_2\mu_1\mu_2^2 + 9k_1k_2\mu_2^2\big\} \\ &\quad \times \big\{(\mu_1+\mu_2)(\mu_2+3k_2+3\mu_1)\big(3\mu_1k_1+\mu_1^2+3k_1\mu_2+7\mu_1\mu_2\big)\big\}^{-1}c + \mathrm{O}(c^2). \end{aligned} \tag{18}$$

Here, we have reported, for sake of brevity, only the first order approximations instead of the complete expressions, which are very complicated. These are relations that hold on for a fibrous material where each fibre or rod is randomly oriented in the space. In other words they may be applied to fibre-reinforced plastics or polymers (with random three dimensional arrangement).

Finally, adopting the characteristic Eshelby tensor of the planar structures we obtain the following relationships for the equivalent elastic moduli of the overall heterogeneous medium:

$$\begin{aligned} k_{eq} &= \frac{k_1(4\mu_2+3k_2)+4c\mu_2(k_2-k_1)}{4\mu_2+3k_2-3c(k_2-k_1)} = k_1+\frac{4\mu_2+3k_1}{4\mu_2+3k_2}(k_2-k_1)c+\mathrm{O}(c^2),\\ \mu_{eq} &= \mu_2\frac{5\mu_1(4\mu_2+3k_2)+c(\mu_2-\mu_1)(9k_2+8\mu_2)}{5\mu_2(4\mu_2+3k_2)-6c(\mu_2-\mu_1)(k_2+2\mu_2)}\\ &= \mu_1+\frac{1}{5}\frac{(9k_2\mu_2+8\mu_2^2+12\mu_1\mu_2+6\mu_1k_2)(\mu_2-\mu_1)}{\mu_2(3k_2+4\mu_2)}c+\mathrm{O}(c^2). \end{aligned} \tag{19}$$

We remember that this latter result is valid for $e=0$ and for any value of g: it describes the overall behaviour of a random embedding of slabs or sheets in a given matrix. Moreover, the result given in Eq. (19) is particularly important because, as we will show in the sequel, it is correct for any value of the volume fraction of the sheets and not only for strongly diluted mixtures. In fact, if we apply the differential scheme to Eq. (19) we obtain, as result, Eq. (19) itself (see next section for details).

At this stage, we have obtained, from the general theory, three mixing rules for spherical, cylindrical and planar objects randomly embedded in the homogeneous matrix. Now, we may obtain many other limiting cases taking into account special kind of the media involved in the mixture definition. In particular, both the homogeneous matrix and the inclusions may assume the role of the vacuum or of a completely rigid medium (that do not allow any deformation). So, we generate an interesting casuistry, which is summarised in Table I and which represents a useful tool for practical applications.

3.1. Special media in dispersions of spheres

We begin with dispersions of spheres: if the spherical inclusions are pores (voids) the material (2) has zero moduli, i.e., $k_2=0$ and $\mu_2=0$ (or $E_2=0$). In the present case, Eq. (17) may be simply written in terms of the Young modulus and the Poisson ratio which are related to the bulk and shear moduli by means of the standard relations:

$$E=\frac{9k\mu}{\mu+3k};\ \nu=\frac{3k-2\mu}{2(\mu+3k)}\quad\Leftrightarrow\quad\mu=\frac{E}{2(1+\nu)};\quad k=\frac{E}{3(1-2\nu)}. \tag{20}$$

Therefore, using $E_2=0$ combined with Eqs. (17) and (20) we obtain the following relationships for a material with spherical pores:

$$\begin{aligned} \nu_{eq} &= \frac{5\nu_1^2c-3c+2c\nu_1-14\nu_1+10\nu_1^2}{15\nu_1^2c-13c+2c\nu_1-14+10\nu_1} = \nu_1-\frac{3}{2}\frac{(1-5\nu_1)(1-\nu_1^2)}{5\nu_1-7}c+\mathrm{O}(c^2),\\ E_{eq} &= \frac{2(1-c)(5\nu_1-7)E_1}{15\nu_1^2c-13c+2c\nu_1-14+10\nu_1} = E_1-\frac{3}{2}\frac{E_1(5\nu_1+9)(\nu_1-1)}{5\nu_1-7}c+\mathrm{O}(c^2). \end{aligned} \tag{21}$$

We note that the Poisson ratio of the porous material depends only on the matrix Poisson ratio and on the porosity.

If the spherical inclusions are completely rigid the strain inside the inclusions must be zero and therefore we let $\mu_2\to\infty$: this condition completely describes a solid with no elastic deformations allowed. As before, starting from Eq. (17) and letting $\mu_2\to\infty$, we observe that the equation for the bulk modulus remains unchanged and the other one, for the shear modulus becomes $\mu_{eq}=\mu_1+5\mu_1(4\mu_1+3k_1)c/6k_1+\mathrm{O}(c^2)$. For instance, in this first order approximation, if we let $k_1\to\infty$ we obtain the very simple relation $\mu_{eq}\approx\mu_1(1+5/2c)$ which describes a mixture of inelastic spheres in a matrix with $k_1\to\infty$. This is the elastic version of the well-known Einstein (1906) result for the viscosity of a dispersion of rigid spheres in a viscous incompressible fluid (in fluid theory $k\to\infty$ means incompressibility of the liquid and μ behaves as the viscosity): if very small rigid spheres are suspended in a liquid, the viscosity is thereby increased by a fraction which is equal to $5/2$ times the total volume of the spheres suspended in a unit volume, provided that this total volume is very small. Anyway, converting our relations to Young modulus and Poisson ratio, Eq. (17) with $\mu_2\to\infty$ (or equivalently $E_2\to\infty$) leads to the result for spherical rigid inclusions:

$$\begin{aligned} \nu_{eq} &= \frac{c(10\nu_1^2-11\nu_1+3)+(8\nu_1-10\nu_1^2)}{30\nu_1^2c+13c-41c\nu_1+8-10\nu_1} = \nu_1+\frac{3}{2}\frac{(1-5\nu_1)(1-2\nu_1)(\nu_1-1)}{5\nu_1-4}c+\mathrm{O}(c^2),\\ E_{eq} &= E_1\frac{2(7-19\nu_1+10\nu_1^2)c^2+(23-50\nu_1+35\nu_1^2)c+(8-2\nu_1-10\nu_1^2)}{(-13+28\nu_1+11\nu_1^2-30\nu_1^3)c^2+(5-26\nu_1-\nu_1^2+30\nu_1^3)c+(8-2\nu_1-10\nu_1^2)}\\ &= E_1+3E_1\frac{(\nu_1-1)(5\nu_1^2-\nu_1+3)}{(\nu_1+1)(5\nu_1-4)}c+\mathrm{O}(c^2). \end{aligned} \tag{22}$$

Once again, ν_{eq} depends only on ν_1 and c.

Other two cases are considered but they are quite trivial: if $E_1 = 0$ we have the vacuum as matrix medium and so we obtain a not connected structure with $E_{eq} = 0$; if $E_1 \to \infty$ we deal with a structure completely blocked by the connected rigid matrix and thus $E_{eq} \to \infty$.

3.2. Special media in dispersions of cylinders

Now we are dealing with a fibrous material formed by two different phases. If the medium of the fibres or cylinders is the vacuum we let $k_2 = 0$ and $\mu_2 = 0$ in Eq. (18), obtaining a result in terms of the Young modulus and Poisson ratio:

$$\begin{aligned} \nu_{eq} &= \frac{3c\nu_1 + 8c\nu_1^2 - 5c - 15\nu_1}{16\nu_1^2 c - 20c - 4c\nu_1 - 15} = \nu_1 + \frac{1}{15}(1+\nu_1)\left(16\nu_1^2 - 28\nu_1 + 5\right)c + \mathrm{O}\left(c^2\right), \\ E_{eq} &= \frac{15(1-c)E_1}{15 + 20c + 4c\nu_1 - 16c\nu_1^2} = E_1 + \frac{1}{15}E_1\left(16\nu_1^2 - 4\nu_1 - 35\right)c + \mathrm{O}\left(c^2\right). \end{aligned} \tag{23}$$

The latter result represents the characterisation of a fibrous porous medium (with randomly oriented cylindrical voids or pipes). Eq. (23) represents the cylindrical counterpart of the spherical relation, Eq. (21), for porous materials.

If the cylindrical inclusions become rigid ($E_2 \to \infty$) the whole structure remains blocked by the network of rigid rods independently on the elasticity of the matrix medium and so $E_{eq} \to \infty$ (this is true because of the infinite length of the inelastic cylinders).

An interesting case is given by letting $E_1 = 0$, i.e., considering the vacuum as matrix: we obtain a random three dimensional grid of elastic rods embedded in air. The corresponding simple relationships follow from Eq. (18):

$$\nu_{eq} = \frac{1}{2}\frac{2c\nu_2 - 3c + 3}{6 - 5c} = \frac{1}{4} + \frac{1}{6}\left(\nu_2 - \frac{1}{4}\right)c + \mathrm{O}\left(c^2\right), \qquad E_{eq} = \frac{cE_2}{6 - 5c} = \frac{1}{6}E_2 c + \mathrm{O}\left(c^2\right). \tag{24}$$

This means that the Poisson ratio of this random grid is approximately equals to $1/4$ for very small radius of the elastic cylinders. To conclude with mixtures of cylinders we analyse the simple case with $E_1 \to \infty$: the whole composite medium remains blocked by the connected rigid matrix and we simply have $E_{eq} \to \infty$.

3.3. Special media in dispersions of sheets

From now on, we will describe mixtures formed by infinite two-dimensional planes randomly embedded in a given homogeneous matrix: the general behaviour is given by Eq. (19). If the second medium (planar inclusions) is the vacuum ($E_2 = 0$) the sheets generates a not connected elastic matrix and the equivalent Young modulus of the whole medium vanishes: $E_{eq} = 0$. If the planar inclusions become rigid ($E_2 \to \infty$) the whole structure remains blocked by the network of rigid sheets independently on the elasticity of the matrix medium and so $E_{eq} \to \infty$. However, the other two following cases are more interesting: if we let $E_1 = 0$ we are considering the vacuum as matrix and therefore we obtain a random three-dimensional grid of elastic slabs or sheets embedded in air. The corresponding equivalent moduli follow from Eq. (19) combined with the assumption $E_1 = 0$:

$$\begin{aligned} \nu_{eq} &= \frac{3 + 5\nu_2^2 c - 3c + 2c\nu_2 + 12\nu_2 - 15\nu_2^2}{15\nu_2^2 c - 15\nu_2^2 + 2c\nu_2 - 12\nu_2 - 13c + 27} = \frac{5\nu_2 + 1}{5\nu_2 + 9} + \frac{2}{3}\frac{(5\nu_2 - 7)(5\nu_2 - 1)(\nu_2 + 1)}{(5\nu_2 + 9)^2(\nu_2 - 1)}c + \mathrm{O}\left(c^2\right), \\ E_{eq} &= \frac{2c(7 - 5\nu_2)E_2}{15\nu_2^2 c - 15\nu_2^2 + 2c\nu_2 - 12\nu_2 - 13c + 27} = \frac{2}{3}\frac{c(7 - 5\nu_2)E_2}{(5\nu_2 + 9)(1 - \nu_2)} + \mathrm{O}\left(c^2\right). \end{aligned} \tag{25}$$

From the biomedical point of view it interesting to note that Eq. (24) and/or (25) may be used to describe the trabecular bone mechanical properties. Trabecular bone is generally characterized as a cellular solid, or foam, consisting of an interconnected network of rods and plates. It is, on the other hand, a porous, sponge-like network of bone material. By neglecting the fact that the structure of trabecular bone adapts to the loading environment and that it is highly asymmetric (for example in femur and tibia in order to support the existing multi-axial state of stress), the linear model, here described, can be applied to characterise this kind of biomaterial.

Finally, the last case considered deals with a rigid matrix $E_1 \to \infty$: the randomly oriented elastic sheets separate rigid parts of the matrix (that remains not connected). In other words, we may think to a random filling of rigid stones separated or cemented by an elastic medium or paste. Eq. (19) where $E_1 \to \infty$ or $\mu_1 \to \infty$ leads to the explicit expressions:

Table 1
Summary of the casuistry obtained by considering special shapes of the inclusions and special media for the phases. Spheres, cylinders and sheets may be used as embedded particles; moreover, both matrix and inclusions may be void or rigid

	Porous inclusions $E_2 \to 0$	Rigid inclusions $E_2 \to \infty$	Porous matrix $E_1 \to 0$	Rigid matrix $E_1 \to \infty$
Spherical inclusions	Dispersion of spherical pores: see Eq. (21) or Eq. (32) for high concentrations	Dispersion of rigid spheres: see Eq. (22) or Eq. (33) for high concentrations	Not connected structure (isolated spheres in air): $E_{eq} \to 0$ ν_{eq} undetermined	Structure blocked by the connected rigid matrix region: $E_{eq} \to \infty$ ν_{eq} undetermined
Randomly oriented cylindrical inclusions or rods	Random network of porous rods in a connected elastic matrix: see Eq. (23) or Eq. (34) for high concentrations	Structure blocked by the connected network of rigid rods: $E_{eq} \to \infty$ ν_{eq} undetermined	Random connected grid of elastic rods embedded in air: see Eq. (24)	Structure blocked by the connected rigid matrix region: $E_{eq} \to \infty$ ν_{eq} undetermined
Randomly oriented planar inclusions or sheets	The porous planar inclusions generates a not connected elastic matrix: $E_{eq} \to 0$ ν_{eq} undetermined	Structure blocked by the connected network of rigid sheets: $E_{eq} \to \infty$ ν_{eq} undetermined	Random connected network of elastic sheets embedded in air: Eq. (25)	Elastic sheets separate rigid parts of matrix (which remains not connected): see Eq. (26)

$$
\begin{aligned}
\nu_{eq} &= \frac{10\nu_2^2 c + 3\nu_2 - 11c\nu_2 + 3c - 3}{30\nu_2^2 c - 30\nu_2^2 - 41c\nu_2 + 51\nu_2 + 13c - 21} = \frac{1}{7 - 10\nu_2} - \frac{2}{3}\frac{(5\nu_2 - 4)(2\nu_2 - 1)(5\nu_2 - 1)}{(10\nu_2 - 7)^2(\nu_2 - 1)}c + \mathrm{O}(c^2),\\
E_{eq} &= \frac{(5c\nu_2 - 15\nu_2 - 7c + 15)(2c - 4c\nu_2 - 3 + 3\nu_2)E_2}{(\nu_2 + 1)(30\nu_2^2 c - 30\nu_2^2 - 41c\nu_2 + 51\nu_2 + 13c - 21)c}\\
&= \frac{15}{c}\frac{(\nu_2 - 1)E_2}{(10\nu_2 - 7)(\nu_2 + 1)} - 2\frac{(50\nu_2^2 - 70\nu_2 + 27)E_2}{(10\nu_2 - 7)^2(\nu_2 + 1)} - \frac{4}{3}\frac{(2\nu_2 - 1)(5\nu_2 - 4)(5\nu_2 - 1)^2 E_2}{(10\nu_2 - 7)^3(\nu_2 + 1)(\nu_2 - 1)}c + \mathrm{O}(c^2).
\end{aligned}
\tag{26}
$$

In the present case, if the volume fraction of the planar elastic sheets vanishes, the whole structure becomes rigid and thus the Young modulus diverges to infinity. So, the first order expansion of E_{eq} (in Eq. (26)) is actually a Laurent series which takes into account the first order pole appearing for $c = 0$. Therefore, the coefficient of the factor $1/c$ is the residue of the function E_{eq} at the point $c = 0$. All the particular results, shown in the present section, have been summed up in Table 1.

It must be underlined that from a merely mathematical standpoint, each theoretical formula of this section may be applied to mixture of viscoelastic materials: in this case each elastic constant, appearing in a relationship, becomes (for linear materials) a complex modulus of the viscolestic medium (formed by elastic constants, viscosities and frequency), which takes into account the creep (strain response to step stress) or relaxation (stress response to step strain). Consequently, as results, the mixing rules give the complex effective moduli of viscoelastic composite materials, which appear as complex dynamic functions of frequency. These results can be easily compared with actual data because the complex effective moduli of viscoelastic materials may be experimentally measured by a number of techniques giving relaxation curve at discrete frequencies (Eyre et al., 2002).

4. Differential effective medium theory

For the sake of simplicity, the relationships described in the previous sections may be recast in the following unified form, for a given shape of the inclusions embedded in the elastic matrix:

$$
\begin{cases} k_{eq} = F(k_1, k_2, \mu_1, \mu_2, c), \\ \mu_{eq} = G(k_1, k_2, \mu_1, \mu_2, c). \end{cases}
\tag{27}
$$

In Eq. (27) constants k_1 and μ_1 are the elastic moduli of the matrix medium, k_2 and μ_2 are the elastic moduli of the inclusions and k_{eq} and μ_{eq} are those of the overall mixture. Functions F and G are given by Eq. (16) for generally shaped ellipsoids and by relations of the previous section for special cases (spheres Eq. (17), cylinders Eq. (18) or planar inclusions Eq. (19)).

The differential procedure is a method to find a second set of mixture relationships considering a first theory describing the composite material (actually functions F and G). This second theory is usually more efficient than the first one even if the mixture is not strongly diluted because it takes into account the interactions among inclusions.

The concentration of dispersed particles in the immediate neighbourhood of a certain particle is taken into account by the use of an integration scheme that was first introduced by Bruggeman (1935). In this scheme the initially low concentration of the embedded particles is gradually increased by infinitesimal additions of dispersed component. In the course of this process the elastic moduli of the medium around a particle slowly changes from $k_1,\ \mu_1$ to $k_{eq},\ \mu_{eq}$, the final moduli of the homogenised system. We start from Eq. (27) for a mixture where c is the volume fraction of the dispersed phase: we consider the unit volume of mixture (1 m^3) and we add a little volume $dc_0 \ll 1\ m^3$ of inclusions.

Therefore, we consider another mixture between a medium with moduli k_{eq} and μ_{eq} (volume equals to 1 m^3) and a second medium $(k_2,\ \mu_2)$ with volume dc_0. In these conditions the volume fraction of the second medium will be $dc_0/(1+dc_0) \approx dc_0$. So, by using the original relations for the mixture we can write: $k_{eq} + dk_{eq} = F(k_{eq}, k_2, \mu_{eq}, \mu_2, dc_0)$ and $\mu_{eq} + d\mu_{eq} = G(k_{eq}, k_2, \mu_{eq}, \mu_2, dc_0)$.

In the final composite material, with the little added volume dc_0 , the matrix (1) will have effective volume $1 - c$ (m^3) and the dispersed medium (2) will have effective volume $c + dc_0$ (m^3).

The initial volume fraction of the second medium is $c/1$ and the final one is $(c + dc_0)/(1 + dc_0)$; so, it follows that the variation of the volume fraction of inclusions obtained by adding the little volume dc_0 is simply given by: $dc = (c+dc_0)/(1+dc_0) - c/1 = dc_0(1-c)/(1+dc_0) \approx dc_0(1-c)$. Therefore, we obtain $k_{eq} + dk_{eq} = F(k_{eq}, k_2, \mu_{eq}, \mu_2, dc/1-c)$ and $\mu_{eq} + d\mu_{eq} = G(k_{eq}, k_2, \mu_{eq}, \mu_2, dc/1-c)$. With a first order expansion we simply obtain:

$$\begin{cases} k_{eq} + dk_{eq} = F\left(k_{eq}, k_2, \mu_{eq}, \mu_2, \dfrac{dc}{1-c}\right) = F(k_{eq}, k_2, \mu_{eq}, \mu_2, 0) + \left.\dfrac{\partial F}{\partial c}\right|_* \dfrac{dc}{1-c}, \\ \mu_{eq} + d\mu_{eq} = G\left(k_{eq}, k_2, \mu_{eq}, \mu_2, \dfrac{dc}{1-c}\right) = G(k_{eq}, k_2, \mu_{eq}, \mu_2, 0) + \left.\dfrac{\partial G}{\partial c}\right|_* \dfrac{dc}{1-c}, \end{cases} \tag{28}$$

where the symbol $*$ means that k_1 and μ_1 must be substituted by k_{eq} and μ_{eq} and the value $c = 0$ is considered inside the partial derivatives. Simplifying Eq. (28) (by recalling the obvious relations: $k_{eq} = F(k_{eq}, k_2, \mu_{eq}, \mu_2, 0)$ and $\mu_{eq} = G(k_{eq}, k_2, \mu_{eq}, \mu_2, 0)$, which hold on for $c = 0$) we obtain the set of differential equations:

$$\begin{cases} \dfrac{dk_{eq}}{dc} = \dfrac{1}{1-c} \left.\dfrac{\partial F}{\partial c}\right|_*, \\ \dfrac{d\mu_{eq}}{dc} = \dfrac{1}{1-c} \left.\dfrac{\partial G}{\partial c}\right|_*. \end{cases} \tag{29}$$

This system, when two functions F and G are given, defines a new couple of functions, which should better describe the mixture even if it is not strongly diluted, taking into consideration, in a certain approximate way, the interactions among different inclusions. The application of this approach to Eq. (16) allows us to write down the following system (it is immediate to identify the differential terms $\partial F/\partial c|_* = (\alpha + 2\beta)(k_2 - k_{eq})$ and $\partial G/\partial c|_* = (\alpha - \beta)(\mu_2 - \mu_{eq})$, which appear in Eq. (29)):

$$\begin{cases} \dfrac{dk_{eq}}{dc} = \dfrac{(\alpha + 2\beta)(k_2 - k_{eq})}{1-c}, \\ \dfrac{d\mu_{eq}}{dc} = \dfrac{(\alpha - \beta)(\mu_2 - \mu_{eq})}{1-c}, \end{cases} \tag{30}$$

where α and β are calculated with $k_1 = k_{eq}$ and $\mu_1 = \mu_{eq}$. This is the standard differential technique and from the mathematical point of view this approach is perfectly equivalent to those appearing in previous literature: Bruggeman (1935), Laughlin (1977) and Norris (1985). The improvement introduced in Eq. (30), as we will show in the sequel, is given by the fact that the knowledge of the coefficients α and β in closed form allows us to write down and solve the set of differential equations in many cases of practical and technological interest. Moreover, this effective medium theory, as formulated by Eq. (30), allows us to analyse dispersions of generally shaped (arbitrary eccentricities) randomly oriented ellipsoids and not only mixtures of spheres or other specific object. A comparison between the strongly diluted characterisation, explained in the first section of the work, and the differential scheme has been shown in Figs. 3 and 4; Fig. 3 deals with the non-differential theory (Eq. (16)) and Fig. 4 has been obtained by solving the differential Eq. (30) with numerical integration. In these plots, to simplify the problem we take into account randomly oriented ellipsoids of rotation: in this case only one eccentricity describes their shape. We define ξ as the ratio between the lengths of the two different axes of the ellipsoids (ratio between the longer and the shorter axis of the embedded ellipsoids): $\xi > 1$ for prolate ellipsoids (of ovary or elongated form) and $\xi < 1$ for oblate ellipsoids (of planetary or flattened form). Therefore, in Figs. 3 and 4, we have plotted the behaviour of the elastic moduli versus the volume fraction c of the inclusions and the decimal logarithm of the parameter ξ. As far as $k_{eq},\ \mu_{eq}$ and E_{eq} are concerned, we may observe that the valley in correspondence to spherical particles is narrower in the differential scheme than in the standard Eq. (16). The behaviour of heterogeneous materials with planar structure ($\log_{10}\xi \to -\infty$) remains unchanged by adopting the differential procedure. Moreover, the characterisation of materials with randomly oriented cylinders ($\log_{10}\xi \to +\infty$) is sensibly varied

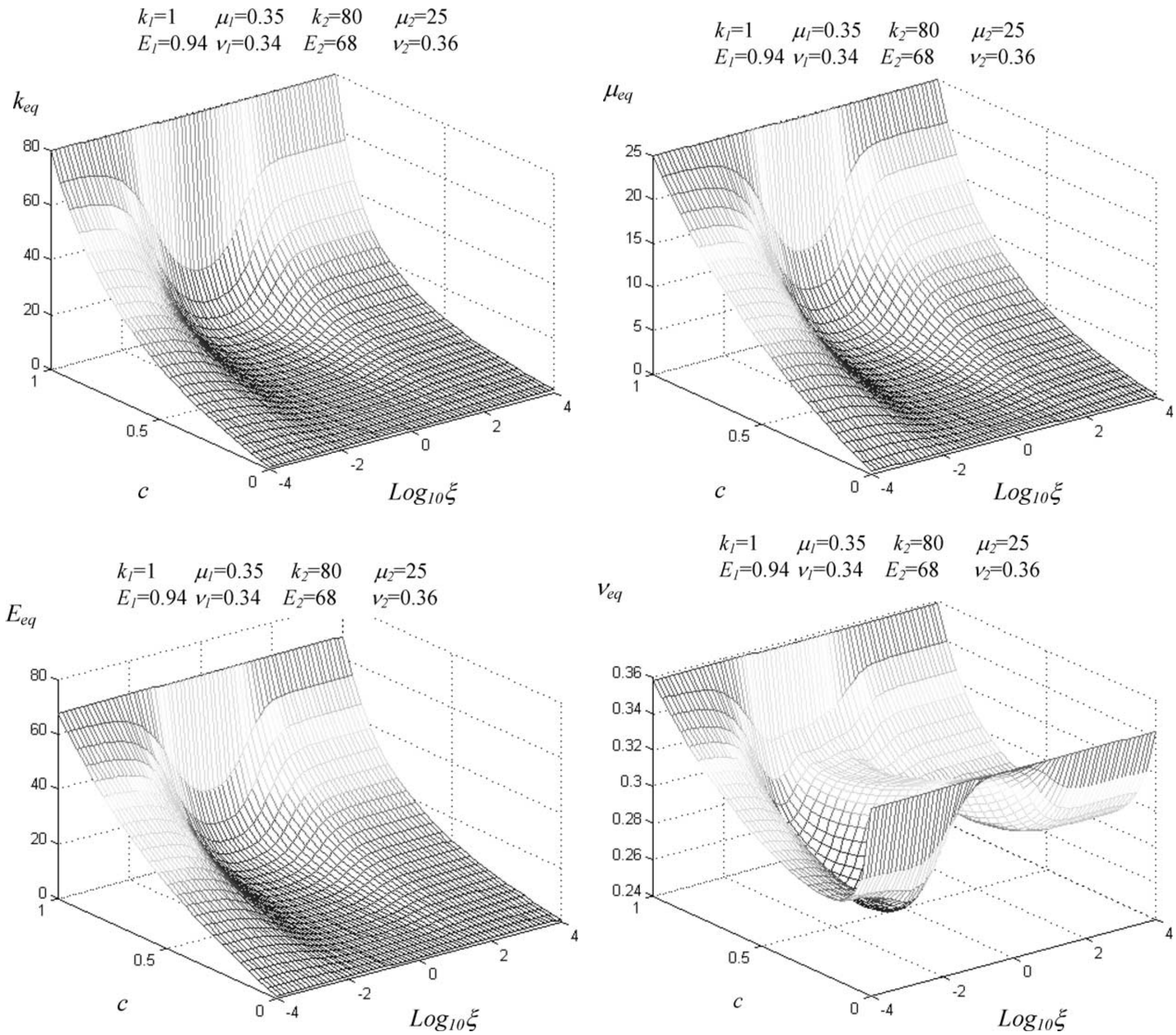


Fig. 3. Results obtained for a mixture of ellipsoids of rotation described by the first procedure (Eq. (16)), holding on for very diluted dispersions. The elastic moduli (bulk, shear, Young and Poisson constants) are shown versus the eccentricity ξ (ratio between the longer and the shorter axis of the embedded ellipsoids) and the volume fraction c of the inclusions.

with the introduction of the differential scheme. Anyway, the larger quantitative differences between the two approaches are exhibited in the zone of spheroidal particles $\log_{10}\xi \approx 0$. Finally, as far as the Poisson ratio is concerned, we confirm the above stated qualitative properties: the overall Poisson ratio is not a monotonic function of ξ, nor a monotonic function of the volume concentration, being allowed to assume values out of the range between ν_1 and ν_2 (Poisson ratio of the matrix and of the inhomogeneities respectively). For the author knowledge, the Poisson ratio is the sole physical parameter that exhibits this very complex scenario as mixing rule.

From now on, we try to apply the differential method to obtain some explicit relations for different classes of heterogeneous materials. For example, Eqs. (21), (22) and (23) of the previous section, may be generalised to higher values of the volume fractions by means of the following differential scheme:

$$\begin{cases} \nu_{\mathrm{eq}} = F(\nu_1, c) \\ E_{\mathrm{eq}} = G(\nu_1, E_1, c) \end{cases} \quad \Rightarrow \quad \begin{cases} \dfrac{\mathrm{d}\nu_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{1}{1-c} \left.\dfrac{\partial F(\nu_1, c)}{\partial c}\right|_{\nu_1=\nu_{\mathrm{eq}},\ c=0}, \\ \dfrac{\mathrm{d}E_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{1}{1-c} \left.\dfrac{\partial G(\nu_1, E_1, c)}{\partial c}\right|_{\nu_1=\nu_{\mathrm{eq}},\ E_1=E_{\mathrm{eq}},\ c=0}. \end{cases} \tag{31}$$

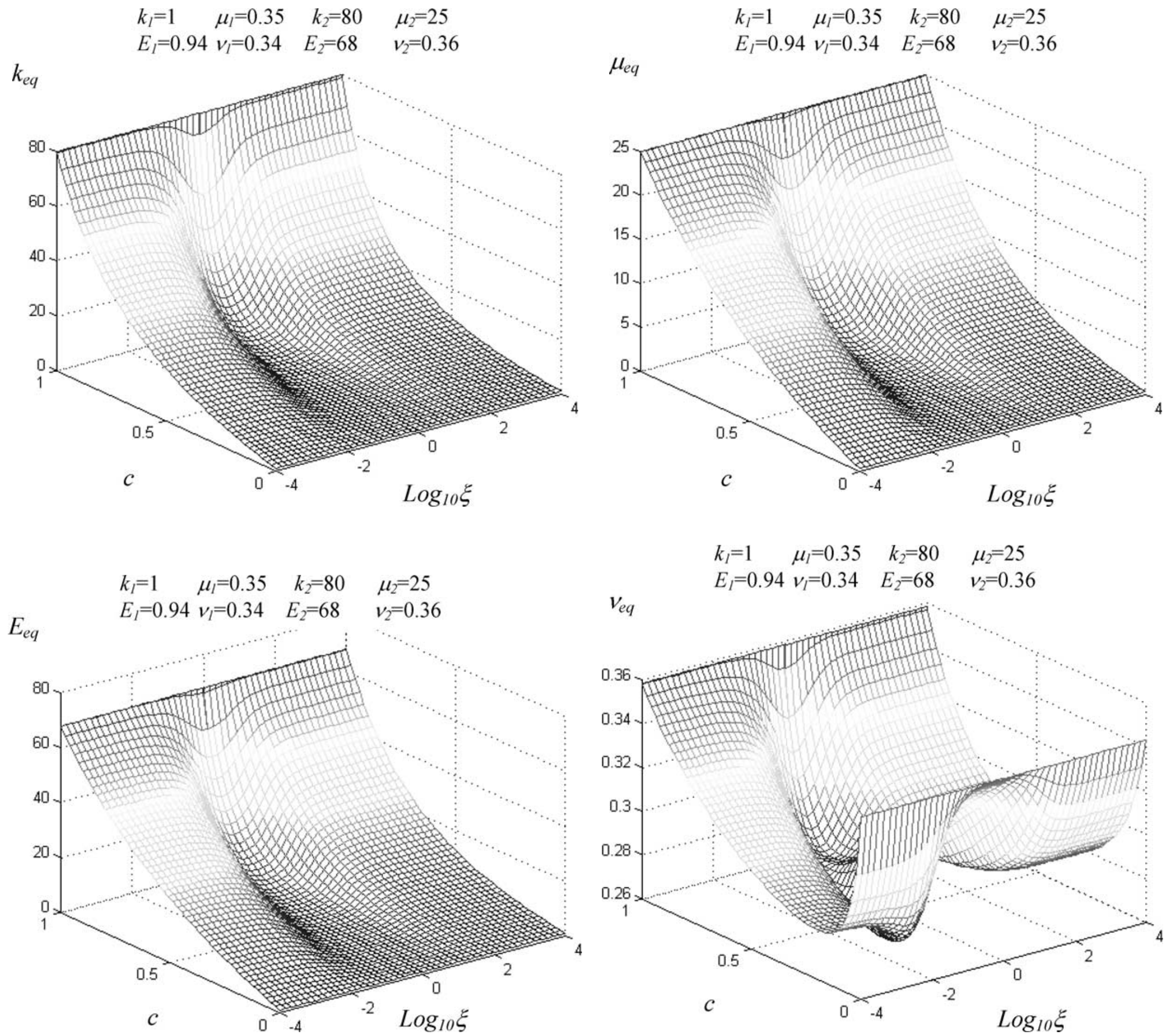


Fig. 4. Results obtained by means of the differential scheme (Eq. (30)) applied to a dispersion of ellipsoids of rotation. The elastic constants (bulk, shear, Young and Poisson) are plotted versus the eccentricity ξ (ratio between the longer and the shorter axis of the embedded ellipsoids) and the volume fraction c with the same assumptions used in Fig. 3 in order to draw a comparison between the theories.

This scheme has the same physical meaning of Eq. (29) but now we are reasoning in terms of the Poisson ratio and the Young modulus. Dealing with dispersions of spheres, we may obtain the following results in closed form. If we are considering the mixture of porous spheres described, for low porosity, by Eq. (21), we can apply the differential procedure, Eq. (31), obtaining the explicit relations:

$$\begin{cases} \left(\dfrac{1-5\nu_{\text{eq}}}{1-5\nu_1}\right)^{5/6}\left(\dfrac{1-\nu_1}{1-\nu_{\text{eq}}}\right)^{1/6}\left(\dfrac{1+\nu_1}{1+\nu_{\text{eq}}}\right)^{2/3} = 1-c, \\ E_{\text{eq}} = E_1\left(\dfrac{1-5\nu_{\text{eq}}}{1-5\nu_1}\right)^{5/3}\left(\dfrac{1+\nu_1}{1+\nu_{\text{eq}}}\right)^{2/3}. \end{cases} \tag{32}$$

In Eq. (32) constants E_1 and ν_1 are elastic moduli of the matrix medium and constants E_{eq} and ν_{eq} are those of the overall porous material with porosity c.

The same procedure may be worked out for the mixture with rigid spheres: this limiting case is described by Eq. (22). Once again, the differential scheme, Eq. (31), may be solved by means of straightforward integration of rational functions, obtaining:

$$\begin{cases} \left(\dfrac{1-5\nu_{\mathrm{eq}}}{1-5\nu_1}\right)^{5/6}\left(\dfrac{1-\nu_1}{1-\nu_{\mathrm{eq}}}\right)^{1/6}\left(\dfrac{1-2\nu_1}{1-2\nu_{\mathrm{eq}}}\right)^{2/3} = 1-c, \\ E_{\mathrm{eq}} = E_1\dfrac{1+\nu_{\mathrm{eq}}}{1+\nu_1}\left(\dfrac{1-2\nu_{\mathrm{eq}}}{1-2\nu_1}\right)^{5/3}\left(\dfrac{1-5\nu_1}{1-5\nu_{\mathrm{eq}}}\right)^{5/3}. \end{cases} \tag{33}$$

Here, constants E_1 and ν_1 are elastic moduli of the matrix medium and constants E_{eq} and ν_{eq} are those of the dispersion of rigid spheres with volume fraction c.

In Eqs. (32) and (33), there is an interesting behaviour of the equivalent Poisson's ratio for high volume fraction of the inclusions: in both models for $c = 1$ the equivalent Poisson's ratio converges to the fixed non-zero value $\nu_0 = 1/5$ irrespective of the matrix Poisson's ratio. This convergent behaviour is exact in two dimensional structures as shown by Day et al. (1992) and by Cherkaev, Lurie and Milton (1992) but unfortunately, at the present state of the research, the available experimental data cannot confirm this qualitative property.

For instance, we can draw some comparisons between Eq. (32) and experimental results. Walsh, Brace and England (1965) made measurements on the compressibility $(1/k_{\mathrm{eq}})$ of a porous glass over a wide range of porosities. Bulk and shear moduli of the pure glass were measured to be $k_1 = 46.3$ GPa and $\mu_1 = 30.5$ GPa, respectively. A comparison between bulk modulus experimentally measured and computed with Eq. (32) is shown in Fig. 5(a). Haglund and Hunter (1973) have studied the Young's modulus of the polycrystalline monoclinic Gd_2O_3. Poisson's ratio and Young's modulus of the pure oxide were measured to be: $E = 150$ GPa and $\nu = 0.29$. In Fig. 5(b) a comparison is drawn between the experimental data and the results given by Eq. (32).

A similar study has been performed for a fibrous porous material: in the present case the strongly diluted characterisation is made by means of Eq. (23). The differential scheme, Eq. (31), can be applied to the Young modulus and the Poisson ratio, obtaining the following expressions, after some long but straightforward computations:

$$\begin{cases} \left(\dfrac{8\nu_{\mathrm{eq}}-7+\sqrt{29}}{8\nu_1-7+\sqrt{29}}\right)^{(15/98)(15/\sqrt{29}+1)}\left(\dfrac{8\nu_1-7-\sqrt{29}}{8\nu_{\mathrm{eq}}-7-\sqrt{29}}\right)^{(15/98)(15/\sqrt{29}-1)}\left(\dfrac{1+\nu_1}{1+\nu_{\mathrm{eq}}}\right)^{15/49} = 1-c, \\ E_{\mathrm{eq}} = E_1\left(\dfrac{8\nu_{\mathrm{eq}}-7+\sqrt{29}}{8\nu_1-7+\sqrt{29}}\right)^{(2/49)(93/\sqrt{29}+16)}\left(\dfrac{8\nu_1-7-\sqrt{29}}{8\nu_{\mathrm{eq}}-7-\sqrt{29}}\right)^{(2/49)(93/\sqrt{29}-16)}\left(\dfrac{1+\nu_1}{1+\nu_{\mathrm{eq}}}\right)^{15/49}. \end{cases} \tag{34}$$

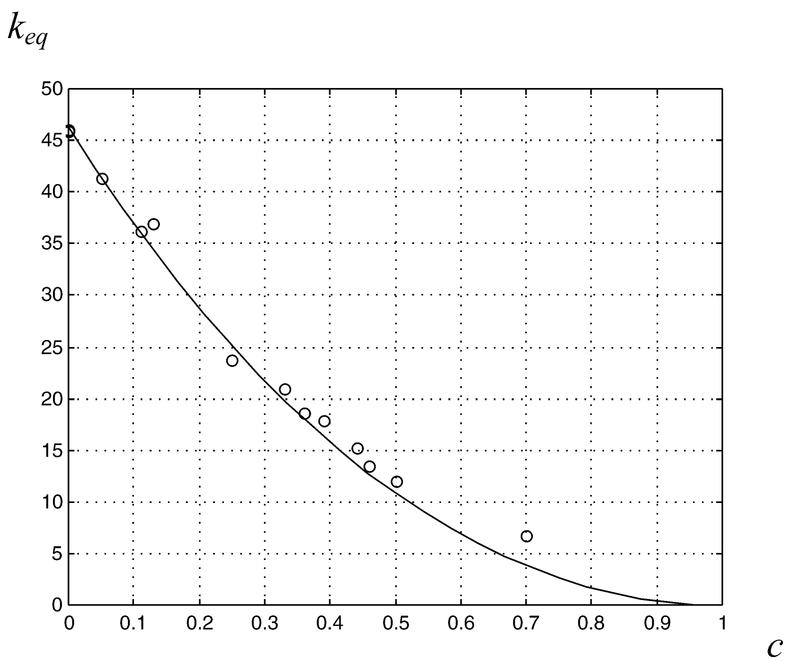
These relations, describing a fibrous porous material for any value of the porosity c, have a particular behaviour very similar to that of Eq. (32) for spherical pores. When the value of the porosity c approaches unity, the Poisson ratio of the composite material converges to the fixed value $\nu_0 = (7-\sqrt{29})/8 \cong 0.2018\ldots$ independently on the value of Poisson ratio of the matrix.

This convergent behaviour of the Poisson ratio, here observed for spherical and cylindrical voids, is actually a peculiarity exhibited for any shape of the pores. We numerically verify the property as follows: from Eq. (5), for porous materials (assuming zero stiffness for the inclusions) we obtain $\hat{\mathbf{A}} = \{\mathbf{I} - \widehat{\mathbf{S}}\}^{-1}$; then we compute the coefficients α and β (Eq. (12)), which depend only on the matrix Poisson ratio and on the eccentricities of the pores. Therefore, we may apply the differential scheme in terms of the Young modulus and the Poisson ratio:

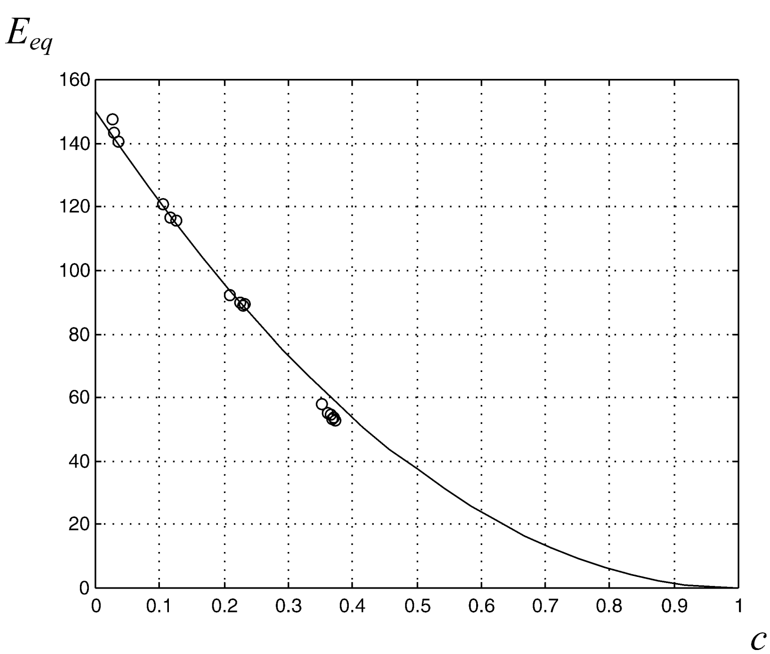
$$\begin{cases} \dfrac{\mathrm{d}E_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{E_{\mathrm{eq}}}{1-c}\left[2\beta(\nu_{\mathrm{eq}})\nu_{\mathrm{eq}} - \alpha(\nu_{\mathrm{eq}})\right], \\ \dfrac{\mathrm{d}\nu_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{\beta(\nu_{\mathrm{eq}})}{1-c}(1+\nu_{\mathrm{eq}})(2\nu_{\mathrm{eq}}-1). \end{cases} \tag{35}$$

The above stated system derives from Eq. (30) when standard transformations between elastic moduli are applied. We have solved Eq. (35) for ellipsoids of rotation. In Fig. 6 elastic moduli are represented for $E_1 = 1$ and $\nu_1 = 0.1$ versus the volume fraction of pores (porosity) and eccentricity ξ (ratio between the longer and the shorter axis of the embedded ellipsoids of rotation). There is a universal behaviour of the Poisson ratio for porous materials (when $c = 1$), which do not depend on the matrix properties. In Fig. 7 the universal function $\nu_0 = \lim_{c\to 1}\nu_{\mathrm{eq}}$ is plotted as function of ξ. There, we may identify the characteristic values for spheres and cylinders: $\nu_0 = 1/5$ for spherical voids and $\nu_0 = (7-\sqrt{29})/8 \cong 0.2018\ldots$ for porous materials with cylindrical randomly oriented voids. Moreover, for planar pores we have: $\lim_{\xi\to 0}\nu_0 = \lim_{\xi\to 0}\lim_{c\to 1}\nu_{\mathrm{eq}} = 0$.

Finally, a particular application of the differential scheme may be performed to analyse the behaviour of composite materials formed by planar inclusions or sheets of medium (2) inserted in the matrix (1). We try to apply the general differential method, described by Eq. (29), to the characterisation of this kind of mixture, which is modelled by Eq. (19). By identifying the pertinent



(a)



(b)

Fig. 5. (a) Bulk modulus k_{eq} in GPa of porous P-311 glass (circles) measured at room temperature compared with data obtained by Eq. (32) (solid line) for different values of the porosity c. Bulk and shear moduli of the pure glass were measured to be $k = 46.3$ GPa and $\mu = 30.5$ Gpa ($E = 75$ Gpa and $\nu = 0.23$). (b) Measured Young's modulus E_{eq} (in GPa) of porous oxide Gd_2O_3 (circles) versus porosity c compared with the solution of Eq. (32) (solid line). For pure oxide, elastic moduli were measured to be $E = 150$ GPa and $\nu = 0.29$.

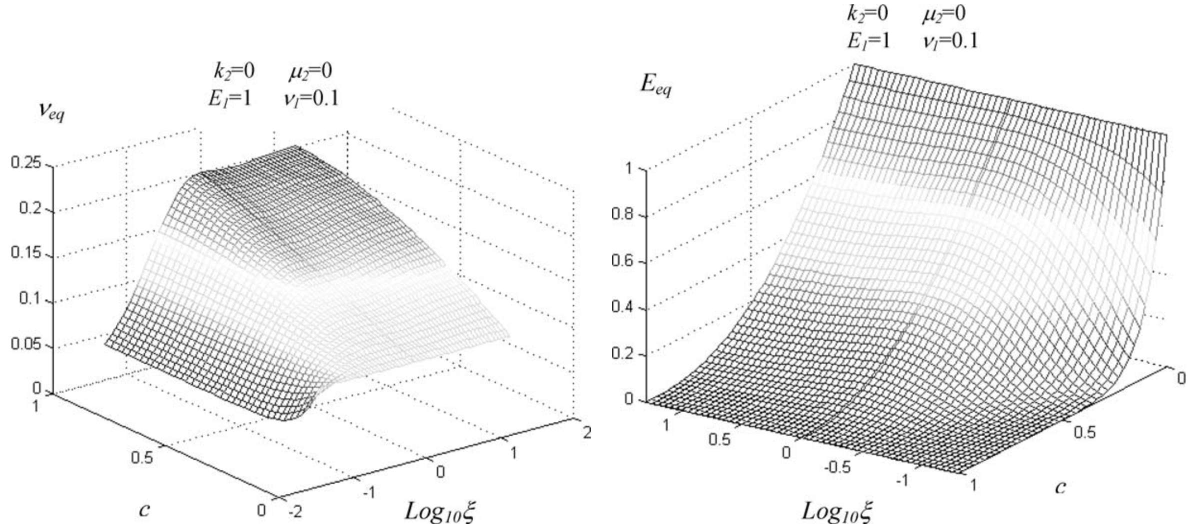


Fig. 6. Application of the differential scheme (Eq. (30)) to the characterisation of porous materials with ellipsoidal (of rotation) voids. Results for the Poisson ratio and the Young modulus versus eccentricity ξ (ratio between the longer and the shorter axis of the embedded ellipsoids) and porosity c are obtained with a matrix described by $E_1 = 1$ and $\nu_1 = 0.1$. For $c \to 1$ we may observe the universal behaviour of the Poisson ratio, which is represented in Fig. 7.

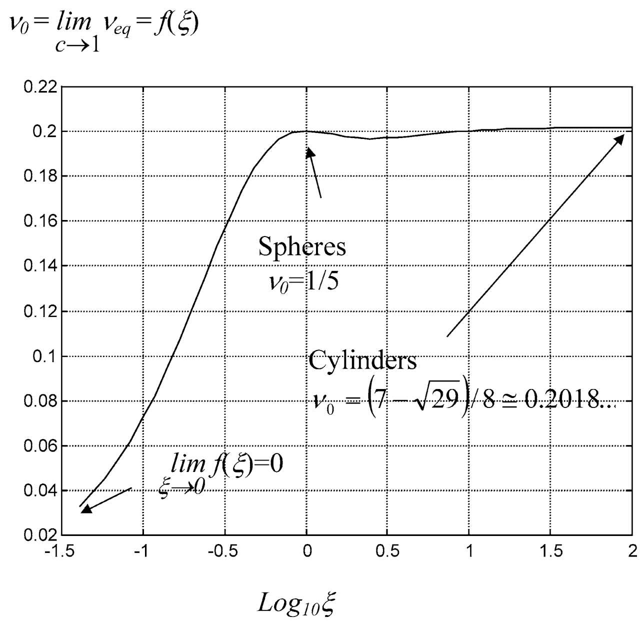


Fig. 7. Universal behaviour of the Poisson ratio for porous materials with high value of the volume fraction (porosity). When $c \to 1$ the Poisson ratio depends only on the eccentricity ξ (ratio between the longer and the shorter axis of the embedded ellipsoids) and not on the matrix elastic properties, as shown in this plot. Characteristic values for spheres and cylinders are clearly evidenced.

functions F and G in Eq. (19) and taking into account the first order approximation we simply obtain the following differential system for the related effective medium theory:

$$\begin{cases} \dfrac{\mathrm{d}k_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{1}{1-c}\dfrac{4\mu_2 + 3k_{\mathrm{eq}}}{4\mu_2 + 3k_2}(k_2 - k_{\mathrm{eq}}), \\ \dfrac{\mathrm{d}\mu_{\mathrm{eq}}}{\mathrm{d}c} = \dfrac{1}{1-c}\dfrac{1}{5}\dfrac{(9k_2\mu_2 + 8\mu_2^2 + 12\mu_{\mathrm{eq}}\mu_2 + 6\mu_{\mathrm{eq}}k_2)(\mu_2 - \mu_{\mathrm{eq}})}{\mu_2(3k_2 + 4\mu_2)}. \end{cases} \tag{36}$$

It is interesting and important to observe that these differential equations are uncoupled and they may be solved separately, obtaining, as results, the relationships appearing in Eq. (19) itself. So, the result stated in Eq. (19) is a fixed point of the differential procedure and therefore it should be correct for any value of the volume fraction of the planar inclusions.

5. Conclusions

We have performed a complete study on the characterisation of dispersions of randomly oriented ellipsoids. The main result of this work is given by an explicit micromechanical averaging technique, which permits to simply analyse, in closed form, the behaviour of randomly oriented objects embedded in a homogeneous matrix. The general theory, developed for diluted mixtures, has been mainly used for two purposes: in the first one we have shown the application of this theory to many limiting cases describing special kind of dispersion (porous material, fibre-reinforced composites, dispersion of flattened inhomogeneities and so on); the latter one represents the analysis of a differential scheme, based on the previous theory, which takes into account any shape of the inclusions and any value of the volume fraction. In both cases we have shown the effects of the microstructure or morphology on the macroscopic effective elastic response of the overall composite material. Moreover, the theory reveals a new interesting behaviour of the Poisson ratio versus the eccentricities of the inclusions and their volume fraction. In particular we have shown that the Poisson ratio of a porous material, for high porosity, depends only on the shape of the voids and not on the elastic response of the matrix.

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