Pergamon

*Int. J. Solids Structures* Vol. 35, Nos. 5–6, pp. 479–486, 1998

0020-7683/98 $19.00 + .00

PII: S0020-7683(97)00029-2

# ON THE STIFFNESS OF COMPOSITES WITH LARGE AMOUNTS OF RIGID PHASE EMBEDDED IN A DUCTILE MATRIX

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(*Received* 3 *September* 1996; *in revised form* 28 *December* 1996)

**Abstract**—A calculation is presented, of the self-consistent genre, to predict the effective isotropic stiffness of a two-phase mixture. The model is valid for high concentrations of rigid equiaxed grains embedded in a power law hardening matrix. A simple formula is derived for the stiffness of such composites as a function of the matrix strain hardening exponent and the concentration. This estimate for the stiffness complements the dilute solution models that exist in the literature. © 1997 Elsevier Science Ltd.

## INTRODUCTION

The effect of the reinforcing phase on the overall stiffness of composites is a subject that has been extensively investigated during the last half century. The particular case where nondeforming equiaxed particles with a volume fraction $c$ are embedded in a ductile matrix has received special attention; usually the particle shape is idealized as being spherical for analytical purposes.

In the dilute solution models the particles are far enough apart that there is virtually no interaction between them. The calculation made by Eshelby (1957), applicable to linear elastic solids, has been widely used on two-phase composites to predict the stiffening effect and Eshelby's calculation has been incorporated into many of the self-consistent estimates, see, for example, Budiansky (1965). If the reinforcing particle is rigid and spherical then Budiansky's result is the same as Eshelby's, i.e., applicable only to dilute solutions where the distance between particles is large enough so that the interaction between particles can be neglected. For spherical particles with diameter $D$, the average spacing $s$ between particles is $s \approx D/\sqrt[3]{c}$ and if, say, for the sake of discussion we demand $s > 5D$ then $c$ must be less than one per cent. In most composites $c$ is larger than this so that there is likely to be mechanical interaction between particles. In spite of this the dilute models are very useful because they provide insight into the asymptotic behaviour of the stiffening effect for a small volume fraction of particles.

In contrast to the non-dilute solutions the mechanical interaction between particles becomes the dominant factor in determining the composite behaviour as $c$ approaches unity and this composition region has received less attention that the dilute region. Figure 1 shows schematically the degree of concentration to which we are referring. Again in this limiting case, where the concentration of rigid grains is sufficiently large so that the main resistance to deformation arises because of hydrostatic constraint on the ductile phase, an analysis to find the stiffening effect of the particles become tractable. Over three decades ago, Drucker (1964) analyzed the mechanical response of such composites. He considered the plane strain deformation of a periodic array of rigid hexagonal grains embedded in an incompressible ductile matrix. Recently, Dryden and Wilkinson (1996) have extended the analysis to three dimensions by considering the creep deformation of a periodic array of rectangular parallelepipeds. One of the features of analyzing a periodic array is that the mechanical properties are anisotropic reflecting the symmetry of the array. On the other hand, a real polycrystal with equiaxed, randomly oriented grains is isotropic with a viscosity $\eta$; its viscosity tensor has spherical symmetry. Finding the relation between the anisotropic

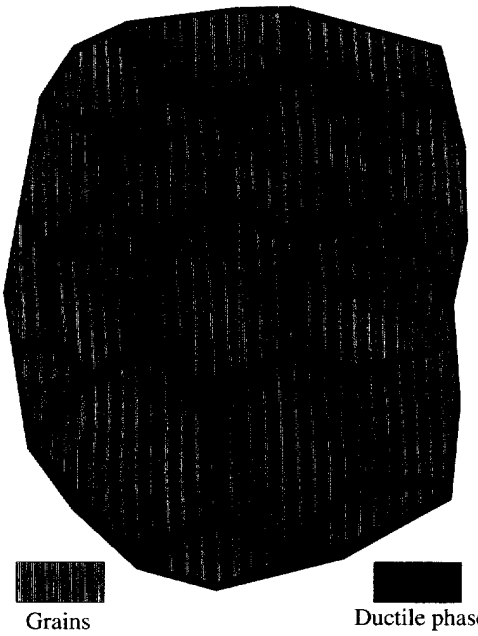


Fig. 1. Schematic diagram of a two-phase composite with a large volume fraction $c$ of equiaxed rigid grains (as shown in the sketch $c \approx 0.7$) embedded in a ductile contiguous matrix phase. The test region enclosed by the dotted line contains an average grain surrounded by a shell of ductile phase. The overall size scale is large enough compared with the grains so that the composite is incompressible and isotropic.

properties of the periodic array and the overall isotropic properties of the composite is similar to the problem is elasticity where the anisotropic elastic properties of the constituent grains are related to the isotropic elastic moduli of the polycrystal. A simple method to obtain upper and lower bounds for the overall properties is to average the stiffness and the compliance tensors over all orientations. (Voigt and Reuss averages), as described by Hirth and Lothe (1982). Because the grains in the periodic array can slide past each other very easily in comparison with being squeezed together (or pulled apart), the bounds obtained by this method are not close. In a real polycrystal the sliding motion is blocked because of its irregular structure so that the upper bound is likely to be more realistic. The upper bound viscosity when compared against the limited amount of experimental data for the effective viscosity of a ceramic containing a small amount of grain boundary phase—although high by a factor of two—does give the correct order of magnitude; the lower bound does not. The estimate subsequently derived in this note shows excellent agreement with the admittedly sparse experimental observations.

In many composites, $0.20 < c < 0.80$, so that the use of neither the dilute nor concentrated models can be completely justified. On an empirical basis it is possible to use a simple cubic spline, given by $\Omega = 3c^2 - 2c^3$, to join a dilute solution model for the stiffness as a function of concentrations, say $D(c)$, to an estimate for the effective properties applicable to a concentrated solution, say $C(c)$, to obtain $E(c) = (1-\Omega)D(c) + \Omega C(c)$. The values of $dE(c)/dc$ are preserved by the spline in each regime. In this note the aim is to obtain a better estimate for $C(c)$: the reinforcing effect of concentrated solutions of equaixed particles embedded in a power law hardening matrix.

Figure 1 shows the test region (enclosed by the dotted line), comprising a rigid grain surrounded by a shell of ductile matrix. We assume in our calculations that the test region is spherical, as shown in Fig. 2, and since there is no size dependence we set the radius to unity. On the dotted line the shear stress is small relative to the normal stress and is taken to be a freely slipping interface. The effective elastic properties of polycrystals (having no boundary phase), with freely-sliding grains was considered by Zener (1941). The analysis was later refined by Ghahremani (1980) who considered a spherical grain, with a freely-slipping interface, embedded in an infinite matrix subjected to a far-field uniform tension.

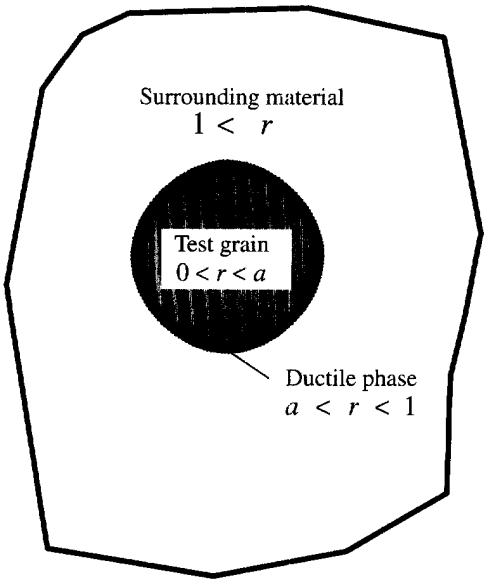


Fig. 2. Dotted line encloses the idealized test region comprising a spherical rigid grain with radius $a$ wrapped in a shell of thickness $\lambda = 1-a$ of ductile matrix phase; $N_0$ is the stiffness of the matrix. The material surrounding the test region is an appropriately chosen continuum with stiffness $N$. The average effective strains in the surrounding material and ductile shell are $\varepsilon_1$ and $\varepsilon_0$ respectively.

There is mechanical interaction between the uniform applied stress field and the stress field arising from the disturbance caused by the slipping; the relation between the slipped and unslipped moduli is found by setting this interaction energy equal to zero. Considering the matrix, in addition to the uniform deformation, the displacement consists of two terms; one decays as $r^{-2}$ and the other as $r^{-4}$. The interaction energy depends only on the $r^{-2}$ term and Dryden (1988) has shown that Ghahremani's expression for the effective elastic stiffness is obtained if the coefficient of this displacement term is set to zero. Moreover, the $r^{-4}$ displacement term causes on average no strain in the matrix. In the analysis that follows we discard the displacement term involving $r^{-2}$ in the surrounding material.

## ANALYSIS

Figure 2 shows the test region comprising of a rigid spherical grain, with radius equal to $a$, enclosed by a spherical shell of ductile matrix in the region $a < r < 1$. The surrounding material outside $r = 1$ is an appropriately chosen continuum with the same mechanical properties as the bulk material and the far-field deformation is uniform.

Overall the composite is isotropic, incompressible and if it has a constitutive equation of the form $S = N\varepsilon^m$ then a potential

$$\mathscr{W}(\varepsilon) = \frac{N}{m+1}\varepsilon^{m+1} \tag{1}$$

which is homogeneous function of degree $m+1$ of the strain components is often used to find the response to multiaxial stress states. The effective strain $\varepsilon = \sqrt{\frac{2}{3}\varepsilon_{ij}\varepsilon_{ij}}$ is an invariant and is equal to the axial strain in pure tension. The effective stress $S = \sqrt{\frac{3}{2}\sigma_{ij}\sigma_{ij}}$ is an invariant of the deviatoric stress components and the stress components are found by taking the partial differentials of $\mathscr{W}$ with respect to the strain

$$\sigma_{ij} = \frac{\partial \mathscr{W}}{\partial \varepsilon_{ij}} = \frac{2}{3}N\varepsilon^{m-1}\varepsilon_{ij}. \tag{2}$$

For the unreinforced matrix the potential has the form

$$\mathscr{W}_o(\varepsilon) = \frac{N_o}{m+1}\varepsilon^{m+1} \tag{3}$$

where the stress components are found as above by differentiating with respect to the strain components. The effective "stiffness" of the composite is equal to $N$ and its relation to the corresponding pure matrix stiffness $N_o$ is what we want to determine.

*Surrounding material*

At a large distance ($r \gg 1$) from the test region, the strain becomes uniform and this incompressible deformation field can be found if the displacement is defined by the gradient of a second order harmonic function. In view of the spherical symmetry we can align the coordinate axes with those of the principal strain directions so that we need to consider only two independent harmonics, say $A(2z^2-x^2-y^2)$ and $B(x^2-y^2)$. Any uniform, incompressible strain field can be formed by using a linear combination of these two harmonics. Solid harmonics of degree equal to $-3$ can be obtained by dividing $2z^2-x^2-y^2$ and $x^2-y^2$ by $r^5$ where $r^2 = x^2+y^2+z^2$. Thus the strain field obtained from

$$\begin{aligned}\chi &= (2z^2-x^2-y^2)\times\left(A+\frac{A'}{r^5}\right)+(x^2-y^2)\times\left(B+\frac{B'}{r^5}\right)\\ &= \left(Ar^2+\frac{A'}{r^3}\right)3\cos^2\phi-1)+\left(Br^2+\frac{B'}{r^3}\right)(\cos^2\theta-\sin^2\theta)\sin^2\phi\end{aligned} \tag{4}$$

where $x = r\sin\phi\cos\theta$, $y = r\sin\phi\sin\theta$ and $z = r\cos\phi$ has no dilational component; the strain components associated with $A'$ and $B'$ decay as $r^{-5}$.

To join the surrounding material to the test region it is convenient to work in spherical coordinates where the displacements are given by

$$u = \frac{\partial\chi}{\partial r}, v = \frac{1}{r}\frac{\partial\chi}{\partial\phi} \quad \text{and} \quad w = \frac{1}{r\sin\phi}\frac{\partial\chi}{\partial\theta}.$$

At the interface between the ductile shell and the surrounding material, i.e., $r = 1$, the shear stress along the interface is much less than the radial stress so that the interface conditions are nearly freely slipping; the shear strains $\gamma_{r\phi}$ and $\gamma_{r\theta}$ vanish if $B = 4B'$ and $A = 4A'$. The radical displacement is then equal to

$$u = \frac{A}{4}\left(8r-\frac{3}{r^4}\right)(3\cos^2\phi-1)+\frac{B}{4}\left(8r-\frac{3}{r^4}\right)(\cos^2\theta-\sin^2\theta)\sin^2\phi. \tag{5}$$

At $r = 1$ the radial displacement is given by

$$\Lambda(\phi,\theta) = \frac{5A}{4}(3\cos^2\phi-1)+\frac{5B}{4}(\cos^2\theta-\sin^2\theta)\sin^2\phi \tag{6}$$

where this is later used as a boundary condition.

The average strain in a given volume can be calculated by using a surface integral. According to Green's theorem, $2\int\varepsilon_{ij}\mathrm{d}V = \int(u_in_j+u_jn_i)\mathrm{d}S$ and within a spherical region of volume $V = \frac{4}{3}\pi R^3$

$$\bar{\varepsilon}_{ij} \equiv \frac{1}{V} \times \frac{1}{2} \int_S \left( \frac{\partial \chi}{\partial x_i} n_j + \frac{\partial \chi}{\partial x_j} n_i \right) \mathrm{d}S = \begin{pmatrix} -2A+2B & 0 & 0 \\ 0 & -2A-2B & 0 \\ 0 & 0 & 4A \end{pmatrix} \tag{7}$$

where $u_i = \partial \chi / \partial x_i$ are the displacement components and $n_i = x_i/R$ are the components of the perpendicular vector on the surface $S$ that encloses the volume $V = \frac{4}{3}\pi R^3$ of the sphere. The average strain $\bar{\varepsilon}_{ij}$ does not depend on either $A'$ or $B'$ and is equal to the far-field uniform strain.

The quantity $\sqrt{\frac{2}{3}\bar{\varepsilon}_{ij}\bar{\varepsilon}_{ij}} = \sqrt{\frac{16}{3}(3A^2+B^2)}$ calculated using the average strain does not account for the stress relaxation caused by the slipping interface. If $\varepsilon_s$ is the average effective strain in the continuum then the energy contained in the unit sphere is $E_s = 4\pi/3 \times N/m+1\ \varepsilon_s^{m-1}$ and according to eqn (2) the average surface traction is given by $\sigma_r = \frac{2}{3} N \varepsilon_s^{m-1}\ \partial u/\partial r$. Using $E_s = 1/m+1 \int \sigma_r \Lambda(\phi, \theta) \mathrm{d}S$ it follows that

$$\varepsilon_s = \sqrt{\frac{1}{2\pi} \times \int_0^{2\pi} \mathrm{d}\theta \int_0^{\pi} \sin\phi\, \mathrm{d}\phi 4\Lambda^2(\phi, \theta)} = \sqrt{\frac{10}{3}(3A^2+B^2)} \tag{8}$$

where $\partial u/\partial r = 4\Lambda(\phi, \theta)$ has been used in the integrand. In the derivation of this expression the average strain has been held fixed so the stress has relaxed as a result of the interface sliding. The quantity of energy

$$E_s = \frac{4\pi}{3} \times \frac{N}{m+1} \left[ \sqrt{\frac{10}{3}(3A^2+B^2)} \right]^{m+1} \tag{9}$$

must be balanced by the deformation within the ductile boundary phase.

*Ductile boundary phase*

In Fig. 2, the ductile phase occupies the boundary region $a < r < 1$ of width equal to $1-a \equiv \lambda$ around the periphery of the rigid test grain where $a \gg \lambda$. At the interface, the radial displacement of the surrounding material which is given in eqn (6) is equated to that of the ductile phase. At $r = a$ the radial displacement is equal to zero so that the average strain in the radial direction is approximately equal to $\Lambda(\phi, \theta)/\lambda$. The average displacement components $V$, $W$ in the $\phi$, $\theta$ directions, respectively can be found using a harmonic potential

$$\psi = \alpha r^2 (3\cos^2\phi - 1) + \beta r^2 \sin^2\phi (\cos^2\theta - \sin^2\theta) \tag{10}$$

where

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left[ r^2 \frac{\partial \psi}{\partial r} \right] = \frac{\Lambda(\phi, \theta)}{\lambda} \quad \text{which gives } \alpha = \frac{5A}{24\lambda}, \beta = \frac{5B}{24\lambda}.$$

Thus

$$V = \frac{1}{r} \frac{\partial \psi}{\partial \phi} = \frac{10}{24\lambda} \times (-3A + B\cos 2\theta) \cos\phi \sin\phi$$

$$W = \frac{1}{r\sin\phi} \frac{\partial \psi}{\partial \theta} = -\frac{10}{24\lambda} \times B \sin 2\theta \sin\phi. \tag{11}$$

These expressions are approximately equivalent to saying that the sum of the strains averaged over $a < r < 1$ must equal zero, i.e. $\bar{\varepsilon}_\phi + \bar{\varepsilon}_\theta + \Lambda(\phi, \theta)/\lambda = 0$.

In view of the fact that $\lambda \gg 1$ it is reasonable to assume that the normal stress components are essentially hydrostatic, i.e., $\sigma_r = \sigma_\phi = \sigma_\theta = \sigma$, and that they do not vary across the boundary thickness. This assumption is made in the theory of elasticity (Fung, 1965), in the analysis of the slow viscous flow of fluid squeezed between two plates (Purday, 1949), and also in Prandtl's solution for rigid-plastic material being squeezed between plates (Kachanov, 1974). Because the displacements $u = v = w = 0$ at $r = a$, the shear strain $\gamma_{\theta\phi}$ is small compared with either $\gamma_{r\phi} \approx \partial v/\partial r$ or $\gamma_{r\theta} \approx \partial w/\partial r$. Mechanical equilibrium requires that

$$\frac{1}{r}\frac{\partial \sigma}{\partial \phi} + \frac{\partial \tau_{r\phi}}{\partial r} = 0$$

$$\frac{1}{r\sin\phi}\frac{\partial \sigma}{\partial \theta} + \frac{\partial \tau_{r\theta}}{\partial r} = 0 \tag{12}$$

so that if $\sigma$ is to be constant across the thickness of the boundary the shear stress must vary linearly with $r$. The matrix which comprises the boundary phase has a power hardening law where the shear stress $\tau$ is proportional $\gamma^m$ so that the displacement is given by

$$v = \frac{n+2}{n+1} \times V \times \left(1 - \frac{\rho^{n+1}}{\lambda^{n+1}}\right)$$

$$w = \frac{n+2}{n+1} \times W \times \left(1 - \frac{\rho^{n+1}}{\lambda^{n+1}}\right) \tag{13}$$

where $n \times m = 1$ and $\rho = 1 - r$ has the range $0 < \rho < \lambda$. The shear strains are then given by

$$\gamma_{r\phi} = \frac{(n+2)V}{\lambda}\frac{\rho^n}{\lambda^n} \quad \text{and} \quad \gamma_{r\theta} = \frac{(n+2)W}{\lambda}\frac{\rho^n}{\lambda^n}$$

so the interface slips freely.

The square of the effective strain is equal to $\frac{1}{3}(\gamma_{r\phi}^2 + \gamma_{r\theta}^2)$ and this is averaged over the surface of the shell to obtain

$$\varepsilon_b = \frac{(n+2)\rho^n}{\sqrt{3}\lambda^{n+1}} \sqrt{\frac{1}{4\pi}\int_0^\pi \sin\phi \, \mathrm{d}\phi \int_0^{2\pi} \mathrm{d}\theta [V^2 + W^2]}$$

$$= \frac{(n+2)\rho^n}{\lambda^{n+2}} \sqrt{\frac{5}{216} \times (3A^2 + B^2)}. \tag{14}$$

The matrix comprising the shell has its potential as given in eqn (3) and therefore the total energy in the shell is equal to

$$E_b = 4\pi \frac{N_o}{m+1} \int_0^\lambda \varepsilon_b^{m-1} \mathrm{d}\rho$$

$$= 4\pi \frac{N_o}{m+1} \frac{(n+2)^m}{\lambda^{2m+1}} \left[\sqrt{\frac{5}{216}(3A^2 + B^2)}\right]^{m-1}. \tag{15}$$

Finally, we equate $E_b$ with the expression for $E_s$, which is given in eqn (9), to obtain

$$\frac{N}{N_o} = \frac{1}{4}\left(\frac{n+2}{12}\right)^m \frac{1}{\lambda^{2m+1}}. \tag{16}$$

This is the quantity of interest in this note; the concentration $c = a^3 \approx 1-3\lambda$ so that eqn (16) is the estimate for $C(c)$ as described in the introduction.

## RESULTS AND DISCUSSION

Since the aim of the calculation is to predict the overall isotropic response of the composite it is of interest to find out whether or not the average radial stress across the interface is continuous. The stress field in the surrounding material, derived using eqn (2) with the deformation field resulting from $\chi$, does not satisfy the equilibrium conditions unless we consider the average stress obtained by replacing $\varepsilon^{m-1}$ with $\varepsilon_s^{m-1}$ and at the interface $\sigma_r = \frac{2}{3}N\varepsilon_s^{m-1}4\Lambda(\phi, \theta)$ where $\partial u/\partial r = 4\Lambda(\phi, \theta)$. The mechanical energy is therefore given by $E_s = 1/(m+1)\int \sigma_r\Lambda(\phi, \theta)\mathrm{d}S = (4\pi/3)N/(m+1)\varepsilon_s^{m+1}$ as given in eqn (9). Equivalent to the expression given in eqn (15) the quantity $E_b = 1/(m+1)\int \sigma\Lambda(\phi, \theta)\mathrm{d}S$ so that imposing the condition $E_b = E_s$ ensures that the average radial stress, i.e., the Fourier coefficient corresponding to $\Lambda(\phi,\theta)$, is balanced at the interface. In the linear case when $m = 1$, the radial stress in the surrounding material $\sigma_r = 2\mu(\partial u/\partial r) = 8\mu\Lambda(\phi, \theta)$, while in the ductile boundary phase it follows from eqns (10–13) that $\sigma = 3\mu_o\psi/\lambda^2 = \mu_o\Lambda(\phi, \theta)/2\lambda^3$ where $\mu_o = N_o/3$ and $\mu = N/3$ are the shear moduli in the matrix and composite. These two expressions for $\sigma_r$ are equal at the interface if $\mu_o = 16\mu\lambda^3$; this is the result given in eqn (16). In the non-linear case it is difficult to evaluate $\sigma$ and the only thing we can say is that on average the normal stress is continuous across the boundary.

It has been assumed that the interface slides freely and the question arises as to how large $c$ must be for this to be a valid assumption. Using eqns (12) and (13) it follows that $\sigma$ depends on $\lambda^{-(1+2m)}$. In contrast, the shear strain associated with the relative sliding between two grains depends on $\lambda^{-1}$ so that the shear stress $\tau$ caused by sliding depends on $\lambda^{-m}$. We have neglected $\tau$ in comparison with $\sigma$ and $\tau/\sigma$ depends on $\lambda^{1+m}$. The volume fraction $c = a^3 = (1-\lambda)^3$ so that if we demand $\lambda < 0.1$ this corresponds to about concentrations greater than 70 per cent.

By analyzing the deformation of a periodic array of cubic grains with Newtonian grain boundary phase, Dryden and Wilkinson (1996) have obtained the constitutive relation $\sigma_{ij} = H_{ijkl}\dot{\varepsilon}_{kl}$, where the fourth order tensor $H_{ijkl}$ has cubic symmetry. Taking the Voigt average as described by Hirth and Lothe (1982) provides an upper bound for the stiffness given by $\eta/\eta_o = 18/5 \times 1/f^3$ where the effective isotropic viscosity is $\eta$, the viscosity of the boundary phase is $\eta_o$ and $f$ is the volume fraction of boundary phase. According to the present calculation, given in eqn (16), by setting $3\lambda = f$ we obtain an expression for the viscosity $\eta/\eta_o = 27/16 \times 1/f^3$ which is about one half the Voigt estimate; as stated in the introduction this lower estimate is in good agreement with experimental results.

Although there are some composites where this model is applicable (such as tungsten carbide particles embedded in ductile cobalt matrix and also in some ceramics where there is a viscous boundary phase), it is also useful insofar as it complements existing models for the reinforcement of less concentrated composites. For instance, Bao *et al.*, (1991) have recently investigated this reinforcement by considering the deformation in an axisymmetric cell, containing matrix and a particle, that is subjected to suitable boundary conditions on its sides. They have also refined the self-consistent calculation made by Duva (1984) to obtain the result $N = N_o(1-c)^{-\alpha}$ where $\alpha \approx 0.39(1-m)+2.5m$ and according to the results of their finite element analysis Duva's expression for $N$ under-estimates the strengthening at non-dilute concentrations of particles. Duva's prediction for the stiffness is also somewhat less than the effect given in eqn (16) which has the stiffness depending on $\lambda^{-2(m+1)}$; at high concentrations $(1-c) \approx 3\lambda$ so that Duva's model predicts strengthening according to $\lambda^{-\alpha}$. Very often, composites have a concentration that is intermediate so that the physics of neither the dilute nor the concentrated type of model is applicable. If we use a model such as Duva's for the composition regime near $c \approx 0$ in conjunction with the behaviour obtained

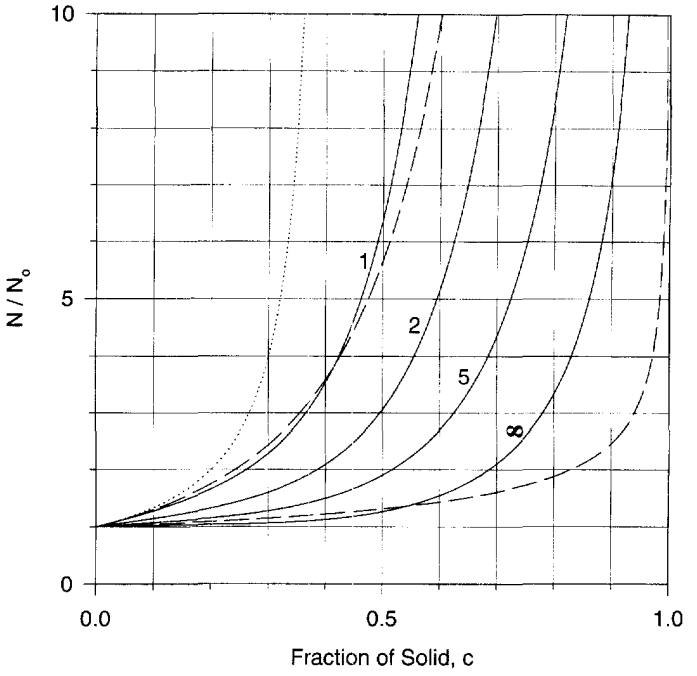


Fig. 3. The change in composite stiffness $N/N_o$ as a function of concentration $c$ for values of $n = 1,2,5$ and $\infty$. The solid lines are obtained using eqn (17). For the sake of comparison, the long dashed lines are Duva's results for values of $n = 1$ and $\infty$; the dotted line is the dilute solution model, valid near $c = 0$, for rigid spheres embedded in a linear elastic matrix and is given by $1/1-2.5c$.

from eqn (16) as $c \to 1$ we can interpolate over the entire composition range $0 < c < 1$ to obtain

$$\frac{N}{N_o} = (1-\Omega) \times (1-c)^{-\alpha} + \Omega \times \frac{1}{4}\left(\frac{n+2}{12}\right)^m \frac{1}{(1-\sqrt[3]{c})^{2m+1}} \tag{17}$$

where $\Omega = 3c^2 - 2c^3$ is the spline mentioned in the introduction. The behaviour of $N/N_o$ is shown in Fig. 3 and as $c \to 1$ the stiffening depends on $\lambda^{-(2m+1)}$.

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