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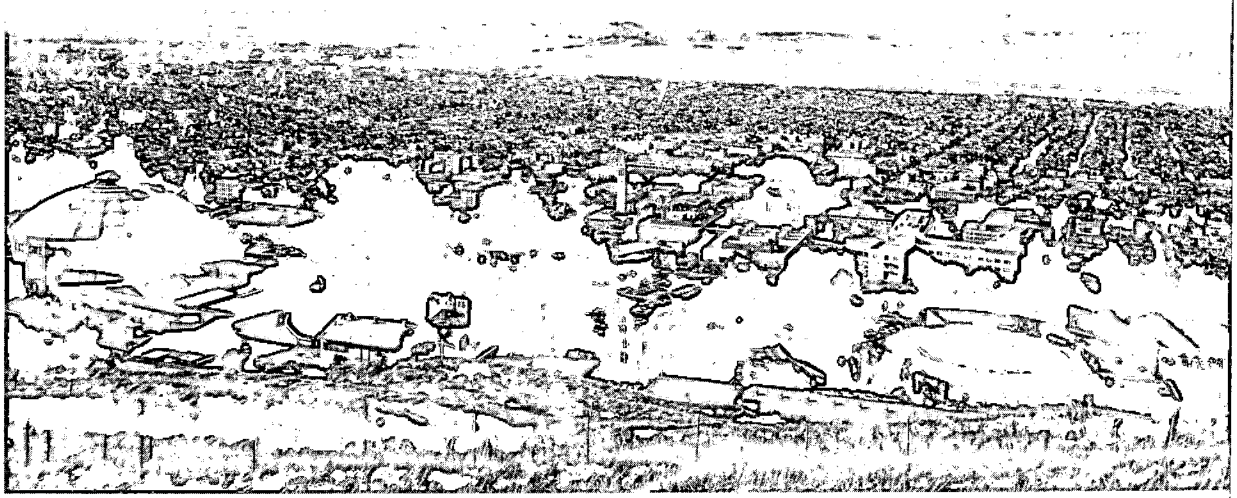

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Elastic Moduli of a Solid Containing Spherical Inclusions

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Abstract

The differential method is used to calculate the elastic moduli of a solid that contains a random distribution of spherical inclusions. Closed-form solutions are obtained for the two limiting cases of rigid inclusions and vacuous pores. These solutions obey the Hashin-Shtrikman bounds, and reduce to the correct values as the inclusion concentration approaches 0 or 1. The predictions are compared with data from the literature, and are shown to be very accurate over wide ranges of the inclusion concentration.

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Introduction

The problem of predicting the effective macroscopic elastic moduli of a microscopically heterogeneous material is an important basic problem in mechanics, and has applications in fields such as composite materials, geophysics, and biomechanics. For the broad class of materials that consist of inclusions dispersed in a continuous matrix, the effective moduli depend on the moduli of the two components, the volumetric concentration of the inclusions, as well as the shape and orientation of the inclusions. Among the many methods that have been proposed to estimate the effective moduli of such materials are the "self-consistent" scheme (Budiansky, 1965; Hill, 1965), the Mori-Tanaka (1973) method, and the "differential" scheme (McLaughlin, 1977; Norris, 1985). In this paper, the differential scheme is applied to materials consisting of isotropic spherical inclusions dispersed in an isotropic matrix. Closed-form solutions are found to the equations in the two limiting cases of rigid inclusions and vacuous pores, and the predictions are compared to data from the literature on various materials.

The differential scheme can be motivated by considering the following "thought experiment". Imagine the addition of a small concentration $\delta\Gamma$ of inclusions to an initially homogeneous matrix material. Since the concentration is dilute, the effect of stress-field interactions between inclusions is negligible, and we can calculate the new elastic moduli using, for example, the method of Eshelby (1957). We then mentally replace this composite material, whose moduli are $M(\delta\Gamma)$, with a homogeneous material having the same moduli. Now imagine the further addition of a small concentration $\delta\Gamma$ of inclusions, and calculate the new moduli $M(2\delta\Gamma)$, etc. If we consider the limit as $\delta\Gamma \rightarrow 0$, we arrive at differential equations that describe the evolution of the effective elastic moduli as functions of the inclusion concentration. Since the inclusions are assumed to be randomly located in the matrix, the second set of inclusions will, for the most part, replace matrix material, but will also replace some of the previously deposited inclusion material. Hence the total inclusion concentration will not simply be an additive function of the amount of inclusions added to the material. As shown by McLaughlin (1977), the total volumetric concentration of the inclusion phase is related to the parameter Γ by $c = 1 - \exp(-\Gamma)$.

If the inclusions are spherical, and both components are isotropic, the two differential equations for the elastic moduli take the form (McLaughlin, 1977; Norris, 1985)

$$\frac{1}{K}\frac{dK}{d\Gamma} = F_K = \frac{(3K + 4G)(K_i - K)}{(3K_i + 4G)K} , \tag{1}$$

$$\frac{1}{G}\frac{dG}{d\Gamma} = F_G = \frac{(15K + 20G)(G_i - G)}{(6K + 12G)G_i + (9K + 8G)G} , \tag{2}$$

where K and G are the bulk and shear moduli, and the subscript i denotes the inclusion phase. The initial conditions for these two equations are that the elastic moduli must be those of the matrix phase when the inclusion concentration is zero, i.e., $K(0) = K_m$ and $G(0) = G_m$. Although these differential equations are coupled and nonlinear, closed-form solutions can be obtained in the two important limiting cases of rigid inclusions and spherical pores. Furthermore, since the functions F_K and F_G are well-behaved, numerical solutions are readily obtainable in all cases.

Rigid Inclusions

In the limiting case of rigid inclusions, the elastic moduli K_i and G_i both approach infinity. Although this double limit may be approached along an infinite number of paths, in any case, (1) and (2) take on the forms

$$\frac{1}{K}\frac{dK}{d\Gamma}=F_K=\left[1+\frac{4G}{3K}\right]=\frac{3(1-\nu)}{(1+\nu)}\ , \tag{3}$$

$$\frac{1}{G}\frac{dG}{d\Gamma}=F_G=\left[\frac{15K+20G}{6K+12G}\right]=\frac{15(1-\nu)}{2(4-5\nu)}\,. \tag{4}$$

Since the right-sides of (3) and (4) are positive, we easily find the (admittedly trivial) result that both of the elastic moduli increase as the inclusion concentration increases. In fact, we can show that the effective elastic moduli both increase beyond bound as the inclusion concentration approaches 100%. To see this, first note that F_K is bounded below by 1, while F_G is bounded below by 15/8. (This second bound is found by examining F_G over the range $0<G/K<3/2$, which corresponds to the admissible range $0<\nu<1/2$.) By the comparison theorem of ordinary differential equations (Corduneanu, 1971, p. 49), we see that K and G grow at least exponentially with Γ, and therefore approach infinity as $\Gamma\to\infty$. However, since the inclusion concentration c is related to Γ by $c=1-e^{-\Gamma}$, we see that $K\to\infty$ and $G\to\infty$ as $c\to 1$. Note also that since $c=1$ is the only singular point of the system (3,4), the moduli will not become infinite at any value of $c<1$.

More information about the qualitative behavior of the solutions to (3) and (4) can be found by forming the differential equation for the Poisson ratio, ν. By differentiating the relation $\nu=(3K-2G)/(6K+2G)$, we first find the general result that

$$\frac{d\nu}{d\Gamma}=\frac{18}{(6K+2G)^2}\left[G\frac{dK}{d\Gamma}-K\frac{dG}{d\Gamma}\right]\,. \tag{5}$$

Combining (5) with (3) and (4) leads to

$$\frac{d\nu}{d\Gamma} = F_\nu = \frac{4(1-\nu)(1-2\nu)(1-5\nu)}{(4-5\nu)} \ . \tag{6}$$

The only roots of $F_\nu(\nu)$ that occur in the physically meaningful range are $\nu=0.5$ and $\nu=0.2$; these are stationary points of (6). Examination of $F_\nu'(\nu)$ shows that $\nu=0.5$ is an unstable stationary point, while $\nu=0.2$ is a stable stationary point, *i.e.*, an *attractor*. A simple way to see this is to note that $F_\nu>0$ when $0<\nu<0.2$, while $F_\nu<0$ when $0.2<\nu<0.5$. Hence rigid inclusions not only increase the elastic moduli, but do so in a manner that drives the Poisson ratio towards the value 0.2. The exception to this behavior occurs if the initial Poisson ratio is exactly equal to 0.5, in which case it will remain equal to 0.5, since $d\nu/d\Gamma$ will equal 0. This is consistent with the effect of the addition of rigid particles into a fluid (which has $G=0$ and hence $\nu=0.5$); this causes an increase in K, but not in G, and so the Poisson ratio remains at 0.5. The behavior of ν as a function of inclusion concentration will be examined in more detail after the solution to (3) and (4) has been presented.

The set of equations (3,4,6), of which only two are independent, could in principle be solved by first integrating (6) using partial fractions (Hashin, 1988), and then substituting the resulting $\nu(c)$ expression into (3) or (4), which would then contain only two variables. However, the equations that would result for K or G would be very unwieldy, and would not be in a form for which partial fraction expansions could be used. Another approach is suggested by the resemblance of the system (3,4) to an autonomous dynamic system, with Γ playing a role analogous to "time". This analogy suggests dividing (3) by (4), and examining the behavior of the system in $\{K,G\}$ "phase space". The differential equation for the "trajectories" is then

$$\frac{dK}{dG} = \frac{2}{5}\frac{K}{G} + \frac{4}{5} \ . \tag{7}$$

Since the right side of (7) depends only on the ratio K/G, we can use the method of Liebnitz (Ince, 1956, p. 18), and define $\lambda = K/G$, in which case (7) takes the form

$$\frac{dG}{G} = \frac{5\,d\lambda}{4-3\lambda}\,. \tag{8}$$

Integration of (8), and use of the initial conditions that specify that $K = K_m$ when $G = G_m$, leads to

$$\frac{G}{G_m} = \left[\frac{(K/G)-(4/3)}{(K_m/G_m)-(4/3)}\right]^{-5/3}. \tag{9}$$

This equation is easily solved for K as a function of G; one more integral is needed to relate the moduli to Γ. By dimensional reasoning, we know that the differential equation for $\lambda = K/G$ as a function of Γ will have a right side that depends only on λ, and not on K or G individually; this suggests deriving and solving the equation for $d\lambda/d\Gamma$. Use of the quotient rule for derivatives leads to

$$\frac{d\lambda}{d\Gamma} = \frac{(4-3\lambda)(4+3\lambda)}{6(2+\lambda)}\,. \tag{10}$$

This equation can be integrated using partial fractions to yield, after some rearrangement of terms,

$$(1-c)^2 = \left[\frac{\lambda-4/3}{\lambda_m-4/3}\right]^{5/3}\left[\frac{\lambda_m+4/3}{\lambda+4/3}\right]^{1/3}. \tag{11}$$

The full solution to the equations is provided by (9) and (11); since (11) is actually a quintic equation for λ, there is little hope of extracting an explicit solution. However, manipulation of (9) and (11), and use of the relationship $\nu = (3K - 2G)/(6K + 2G)$, leads to the following more convenient expressions:

$$\frac{G}{G_m} = \frac{1}{(1-c)^2}\left[\frac{3(1-\nu_m)}{4(1-2\nu_m)-(1-5\nu_m)(G/G_m)^{-0.6}}\right]^{1/3}, \tag{12}$$

$$\frac{K}{G} = \frac{4}{3} - \frac{2(1-5\nu_m)}{3(1-2\nu_m)}\left[\frac{G}{G_m}\right]^{-3/5}. \tag{13}$$

It is easy to verify that as $c \to 1$, $G/G_m \to \infty$ (see (12)), and $K/G \to 4/3$ (see (13)), which corresponds to a Poisson ratio of 0.2. For the special case where $\nu_m = 0.2$, (13) shows that $K/G = 4/3$ for all values of c, and the bracketed term in (12) reduces to unity, so that when $\nu_m = 0.2$,

$$\frac{K}{K_m} = \frac{G}{G_m} = \frac{1}{(1-c)^2}. \tag{14}$$

Another interesting special case is that of a rubber-like material, for which G_m is finite but K_m can be thought of as approaching infinity. For such a material, $\nu_m = 0.5$, in which case (12) reduces to $G/G_m = (1-c)^{-5/2}$, and (13) shows that the bulk modulus remains "infinite", as would be expected. In general, as long as ν_m is not too close to 0.5, three-figure accuracy can be achieved by inserting $G/G_m = (1-c)^{-2}$ into the right side of (12), solving for G/G_m, and then inserting this value into (13) to solve for K/K_m.

Since the solution attains such a simple form for the case $\nu_m = 0.2$, it is convenient to use this value to illustrate the different predictions of the various effective moduli theories. In fact, it so happens that for this particular initial condition, the predictions of the other effective moduli theories take on simple forms, as well. The self-consistent method (Hill, 1965) yields $K/K_m = G/G_m = (1-2c)^{-1}$, while the Hashin-Shtrikman (1961) lower bound on the moduli ratio is $(1+c)/(1-c)$. The Mori-Tanaka method also predicts $(1+c)/(1-c)$ in this case. The various results are plotted in Fig. 1. Note that, as pointed out by Norris (1985), use of the differential method with $\Gamma = c$ instead of $\Gamma = -\ln(1-c)$ would lead to effective normalized moduli of e^{2c}, which would fall slightly below the Hashin-Shtrikman lower bound (see Zimmerman, 1984). McLaughlin (1977) in fact showed, without solving the differential equations, that if we identify Γ with $-\ln(1-c)$, the differential scheme obeys the Hashin-Shtrikman bounds, at least when the inclusions are spherical.

It is also instructive to plot the Poisson ratio as a function of inclusion concentration (Fig. 2). The Poisson ratio can be found from (12) and (13) using the relation $\nu = (3K-2G)/(6K+2G)$. As long as the initial Poisson ratio ν_m is less than 0.5, ν approaches 0.2 as $c \to 1$. The rate of this approach, however, decreases as $\nu_m \to 0.5$. The special case of $\nu_m = 0.5$ can be realized in two physically different ways, each of which is accounted for by (12) and (13). One case, discussed above, is that of a "rubber-like" matrix for which $K_m \to \infty$ while G_m remains finite. The other case is that of a fluid-like matrix, for which $G_m \to 0$ while K_m remains finite. Both cases can be accounted for by first letting $\nu_m \to 0.5$ in (12), and solving for $G/G_m = (1-c)^{-5/2}$. Using this value for G/G_m, (13) can be rearranged to yield $K/K_m = (1-c)^{-1}$. These equations show that K will remain "infinite" for a rubber-like matrix, while G will increase, whereas G will remain zero for a fluid-like matrix, while K will increase. Note that the limiting behavior of the general solution (12) and (13) agrees with that found by Christensen (1990), who integrated (3) and (4) for the special case $\nu_m = 0.5$.

Spherical Pores

Another important limiting case is that of vacuous pores, for which case the elastic moduli of the inclusion phase vanish. The governing differential equations for the effective moduli then take the form

$$\frac{1}{K}\frac{dK}{d\Gamma} = F_K = -\left[1 + \frac{3K}{4G}\right] = \frac{-3(1-\nu)}{2(1-2\nu)} , \tag{15}$$

$$\frac{1}{G}\frac{dG}{d\Gamma} = F_G = -\left[\frac{15K+20G}{9K+8G}\right] = \frac{-15(1-\nu)}{(7-5\nu)} . \tag{16}$$

Following the same reasoning as for the case of rigid inclusions, we see that since $F_K < -1$ and $F_G < -5/3$, both of the effective elastic moduli will approach 0 as the inclusion concentration (*i.e.*, porosity) approaches 100%. The differential equation for the Poisson ratio, which is found by combining (5) with (15) and (16), is

$$\frac{d\nu}{d\Gamma} = F_\nu = \frac{3(1+\nu)(1-\nu)(1-5\nu)}{2(7-5\nu)} . \tag{17}$$

As was the case for rigid inclusions, the value $\nu=0.2$ is a point of attraction for the the Poisson ratio. Unlike that case, however, the value $\nu=0.5$ is not a stationary point. Spherical pores will therefore decrease the elastic moduli of a material in such a way as to cause the Poisson ratio to approach 0.2, regardless of the value of ν_m. The self-consistent equations of Hill (1965) and Budiansky (1965) also predict this behavior for a material with spherical pores (see Rabier, 1989), except that the self-consistent theory predicts that the effective elastic moduli vanish and the effective Poisson ratio reaches 0.20 at a porosity of 50%, rather than 100%.

The equations (15) and (16) can be solved by again working with the differential equation for the trajectories, which in this case takes the form

$$\frac{dK}{K} = \frac{(8\beta+9)d\beta}{3\beta(4\beta-3)}\,, \tag{18}$$

where it has been found to be more convenient to work with $\beta = 1/\lambda = G/K$. Taking into account the proper boundary conditions, the solution to (18) is

$$\frac{K}{K_m} = \frac{\beta_m}{\beta}\left[\frac{\beta-3/4}{\beta_m-3/4}\right]^{5/3}, \tag{19}$$

which can also be written as

$$\frac{G}{K} = \frac{3}{4} + \frac{3(1-5\nu_m)}{4(1+\nu_m)}\left[\frac{G}{G_m}\right]^{3/5}. \tag{20}$$

Substitution of (20) into (16) yields an uncoupled equation for G as a function of Γ:

$$-\frac{d\ln G}{d\Gamma} = \frac{10(1+\nu_m) + 5(1-5\nu_m)(G/G_m)^{3/5}}{5(1+\nu_m) + 2(1-5\nu_m)(G/G_m)^{3/5}}\,. \tag{21}$$

Under the change of variable $u = (G/G_m)^{3/5}$, (21) takes the form

$$\frac{-5}{3u}\frac{du}{d\Gamma} = \frac{10(1+\nu_m)+5(1-5\nu_m)u}{5(1+\nu_m)+2(1-5\nu_m)u}\,. \tag{22}$$

Integration of (22) by partial fraction expansions yields (after the results are re-expressed in terms of G/G_m)

$$\frac{G}{G_m} = (1-c)^2 \left[\frac{2(1+\nu) + (1-5\nu)(G/G_m)^{3/5}}{3(1-\nu)} \right]^{1/3} . \tag{23}$$

This solution, given by (20) and (23), is equivalent to that given by Norris (1985), although in a different form.

For the special case of $\nu_m = 0.2$, (20) shows that $G/K = G_m/K_m = 3/4$, while (23) shows that $G/G_m = (1-c)^2$. For this initial condition, the self-consistent method predicts normalized moduli of $1-2c$ (Hill, 1965), while the Mori-Tanaka (1973) method and the Hashin-Shtrikman (1961) upper bound both give $(1-c)/(1+c)$. These results are plotted in Fig. 3. For other values of ν_m, it is not true that $K/K_m = G/G_m$; however, the relative positions of the three curves are the same, in the sense that K (Mori-Tanaka) > K (Differential) > K (Self-consistent). In general, (20) and (23) can be solved to within 1% accuracy by replacing G/G_m with $(1-c)^2$ on the *right* side of (23), and then evaluating (23) and (20) in turn. The Poisson ratio is plotted in Fig. 4 as a function of the porosity, from the solution given by (20) and (23). The effect of spherical pores on the Poisson ratio is qualitatively similar to the effect of rigid spherical inclusions (Fig. 2), with the exception that $\nu_m = 0.5$ is no longer a special case. Note that since F_ν is a very slowly-varying function of ν (see (17)), the Poisson ratio is nearly a linear function of the porosity.

Comparison with Experimental Data

Although much can be said concerning the motivation behind each of the effective moduli theories, perhaps the best test of these methods is to compare their predictions with the results of carefully conducted experiments. One set of data that is useful in this regard are the bulk modulus measurements of Walsh et al. (1965), who prepared sintered glasses of varying porosities under controlled conditions so as to create nearly spherical pores. Since the function F_K is known to be very insensitive to slight deviations from sphericity (Zimmerman, 1985b), it seems permissable to use this data to test the theoretical predictions for a body with spherical pores. Walsh et al. (1965) extrapolated their data down to zero porosity to find an initial Poisson ratio of 0.193. Using this value of ν_m in (20) and (23), as well as in the self-consistent and Mori-Tanaka expressions (see Christensen, 1990), we can compare the values predicted by these theories to the experimental data. The results plotted in Fig. 5 show that the data points generally fall between the predictions of the Mori-Tanaka and differential schemes, although somewhat closer to those of the latter.

It would be convenient if data existed on the elastic moduli of a material filled with "rigid" spherical inclusions. Examination of (1) and (2) shows that the ratios K_i/K_m and G_i/G_m would have to be on the order of 100 or so for the functions F_K and F_G to reduce to those given in (3) and (4) to within 1%. Furthermore, note that the right sides of (1) and (2) are proportional to the differences $(K-K_i)$ and $(G-G_i)$, and that these differences will *decrease* as the inclusion concentration increases. Hence, while the "rigid filler" limit is useful in that it provides a solvable limiting case that can yield qualitative information, it cannot be used for typical composite materials for which K_i/K_m and G_i/G_m are finite (i.e., about 10 or so, but usually not as large as 100). In this case it is necessary to resort to numerical solutions of (1) and (2) in order to facilitate comparisons with experimental data. In this regard, it is worth noting that it has often been suggested that measurements of the viscosity of a suspension of rigid spheres in a Newtonian fluid be used to test the effective moduli theories (cf., Christensen, 1990), since this viscosity is mathematically analogous to that of the shear modulus of an incompressible matrix loaded with rigid spheres. However, it must be remembered that the problem being attacked by the effective medium theories corresponds to a dispersion that is isotropic and homogeneous on some macroscopic scale. When the viscosity of a suspension is measured in a Couette viscometer, the dispersed spheres tend to migrate away from the walls (Halow and Wills, 1970), creating an inhomogeneous suspension. Similar concentration inhomogeneities occur in other types of viscometers (see Leal, 1980). Since the viscosity is determined by measuring the viscous drag exerted by the fluid on the wall of the viscometer, the value so measured will not correspond to the average volume fraction of the overall suspension. Hence, comparisons of these measured viscosities with effective medium predictions will be problematic.

A useful set of experimental data for a composite material with spherical fillers that are more rigid than the matrix, but not "infinitely" rigid, is that of Hasselman and Fulrath (1965) on a borosilicate glass containing spherical tungsten particles. The particle size of the tungsten was approximately $30\mu m$. (Although the differential scheme is somewhat easier to "justify" under the assumption that there is a hierarchy of particle sizes (cf., Norris, 1985; Christensen, 1990), such an assumption is by no means necessary (Zimmerman, 1984). Furthermore, many potential applications of effective medium theories will be to composites with nearly "monodisperse" inclusion phases.) The elastic moduli of the matrix and inclusion phase for this data were $G_m = 33.6\,\text{GPa}$, $K_m = 44.3\,\text{GPa}$, $G_i = 148.1\,\text{GPa}$, and $K_i = 196.3\,\text{GPa}$. Fig. 6 shows the experimentally determined Young's moduli, for tungsten volume fractions of 0.1, 0.2, 0.3, 0.4, and 0.5, compared with the predictions of the differential scheme, the self-consistent scheme, and the Mori-Tanaka method. In this case, the Mori-Tanaka predictions coincide with the Hashin-Shtrikman *lower* bound (see Benveniste, 1987). Again, the differential scheme fits the data very well. The Mori-Tanaka method is slightly less accurate, while the self-consistent method is the least accurate of these three methods. Note that since the functions F_K and F_G are well-behaved and slowly-varying, equations (1) and (2) are easily integrated if $\ln K$ and $\ln G$ are used as the dependent variables.

Conclusions

Closed-form solutions have been presented for the equations, obtained from the differential scheme, that describe the effective elastic moduli of materials containing spherical pores or rigid spherical inclusions. The predictions lie between those of the Mori-Tanaka method and the self-consistent method, although this may not always be true for other microstructures (cf., Christensen, 1990). In both of these cases, the Poisson ratio approaches 0.2 as the inclusion phase concentration increases. For the general case in which the moduli of the inclusion phase is finite, the equations of the differential scheme are easily integrated numerically. The predictions of the differential scheme compare very well to experimental data from the literature on the bulk modulus of a sintered glass with spherical pores, and on a borosilicate glass containing spherical tungsten particles. The differential scheme also has the advantage of being easily applied to any microstructure for which the Eshelby (1957) energy expressions are available; for example, to materials containing penny-shaped cracks (Salganik, 1973; Zimmerman, 1985a) or rigid-disk inclusions (Norris, 1990).

Acknowledgments

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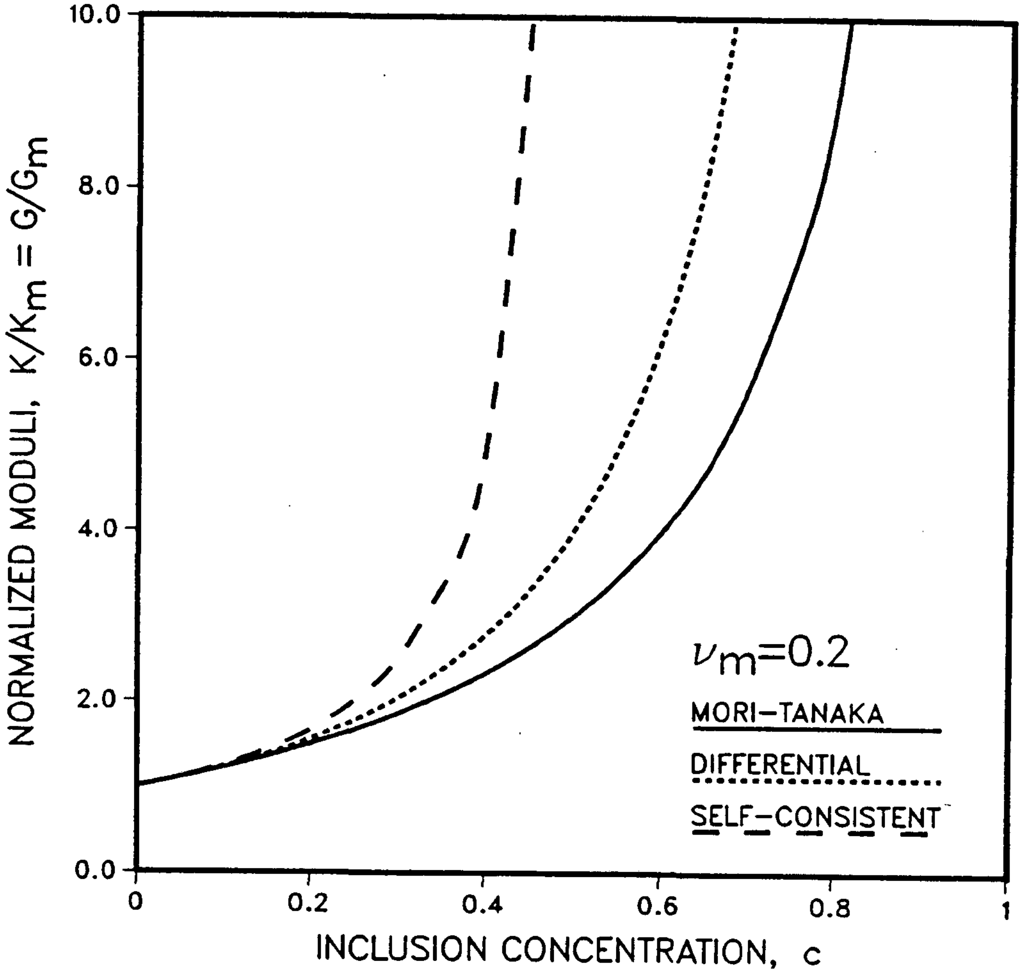

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Fig. 1. Elastic moduli of a material containing rigid spherical inclusions, according to the self-consistent scheme, the Mori-Tanaka scheme, and the differential scheme. The Poisson ratio of the matrix phase is 0.20.

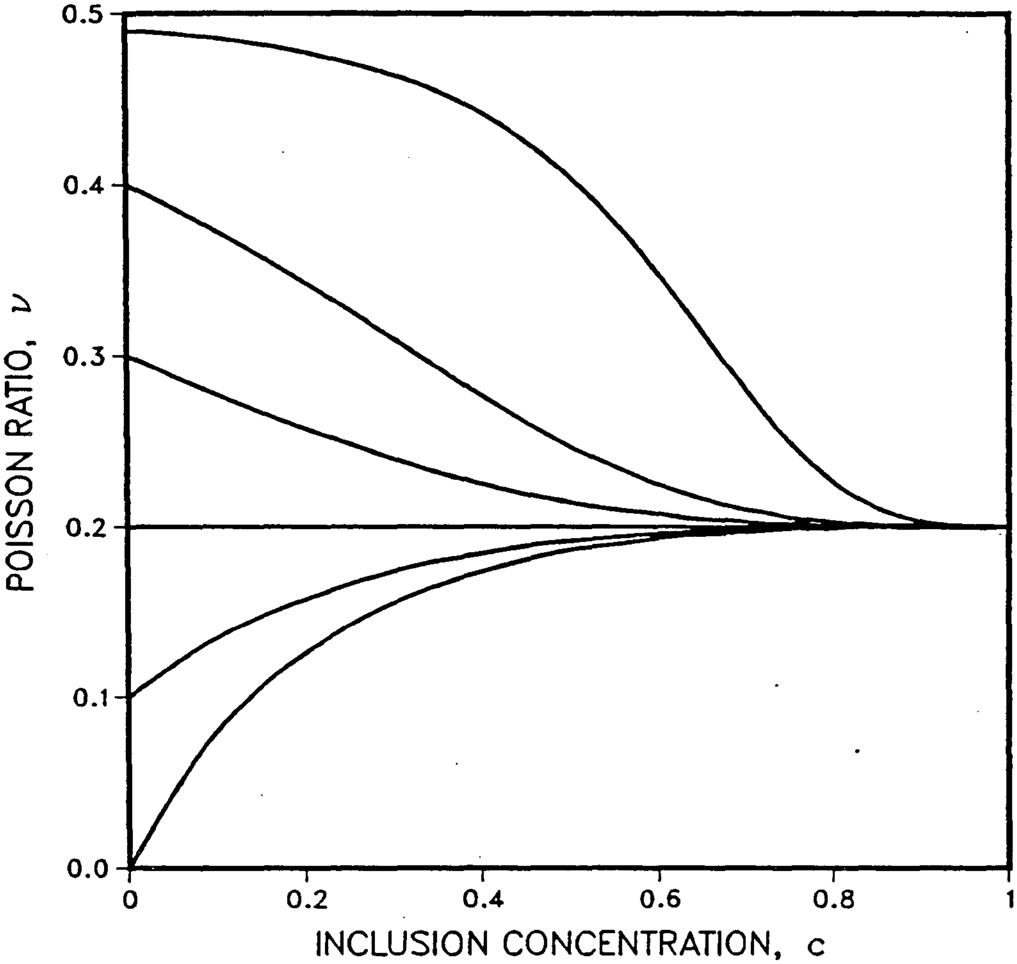

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Fig. 2. Poisson ratio of a material containing rigid spherical inclusions, according to the differential scheme (equations (11) and (12)). The six curves are for ν_m = 0.0, 0.1, 0.2, 0.3, 0.4, and 0.49.

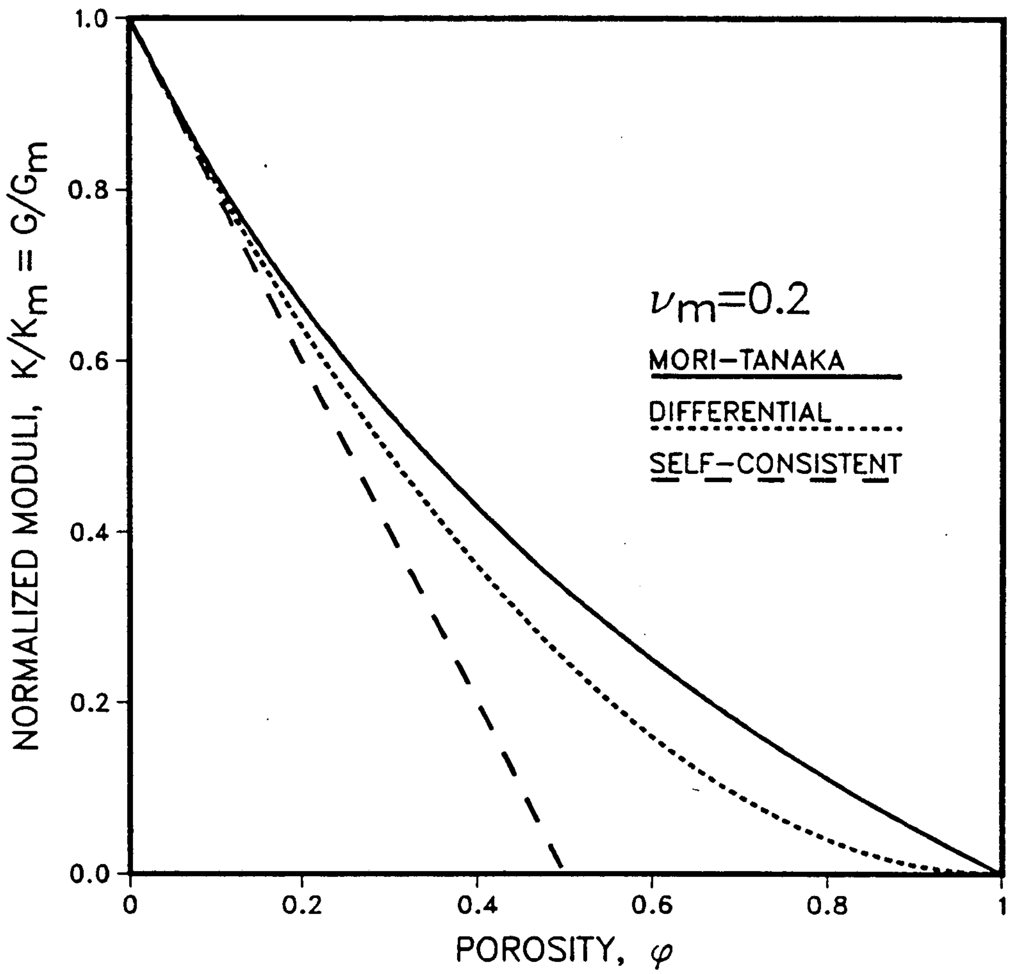

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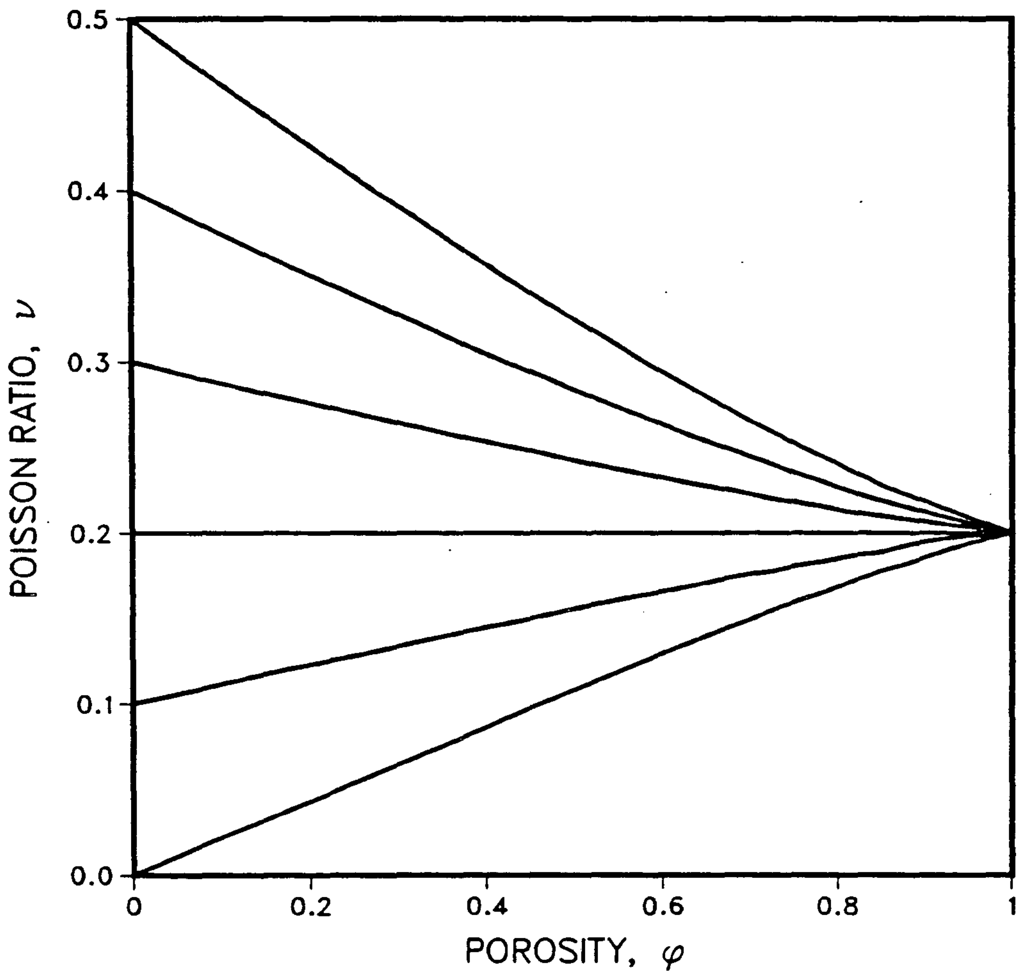
Fig. 3. Elastic moduli of a material containing spherical pores, according to the self-consistent scheme, the Mori-Tanaka scheme, and the differential scheme. The Poisson ratio of the matrix phase is 0.20.



XBL 9012-3984

Fig. 4. Poisson ratio of a material containing spherical pores, according to the differential scheme (equations (20) and (23)). The six curves are for ν_m = 0.0, 0.1, 0.2, 0.3, 0.4, 0.5.

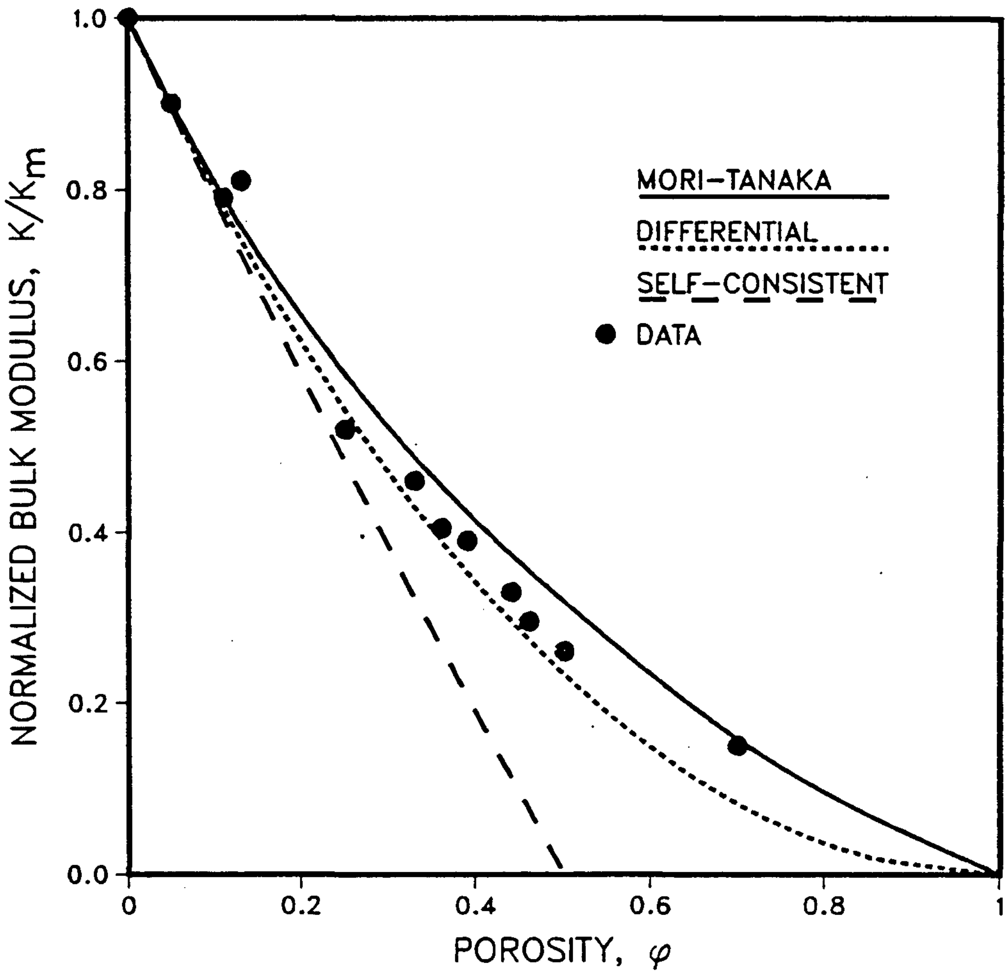


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Fig. 5. Bulk modulus of a sintered glass containing spherical pores, compared with the predictions of the three effective medium theories. The Poisson ratio of the matrix phase is 0.193; data are from Walsh et al. (1965).

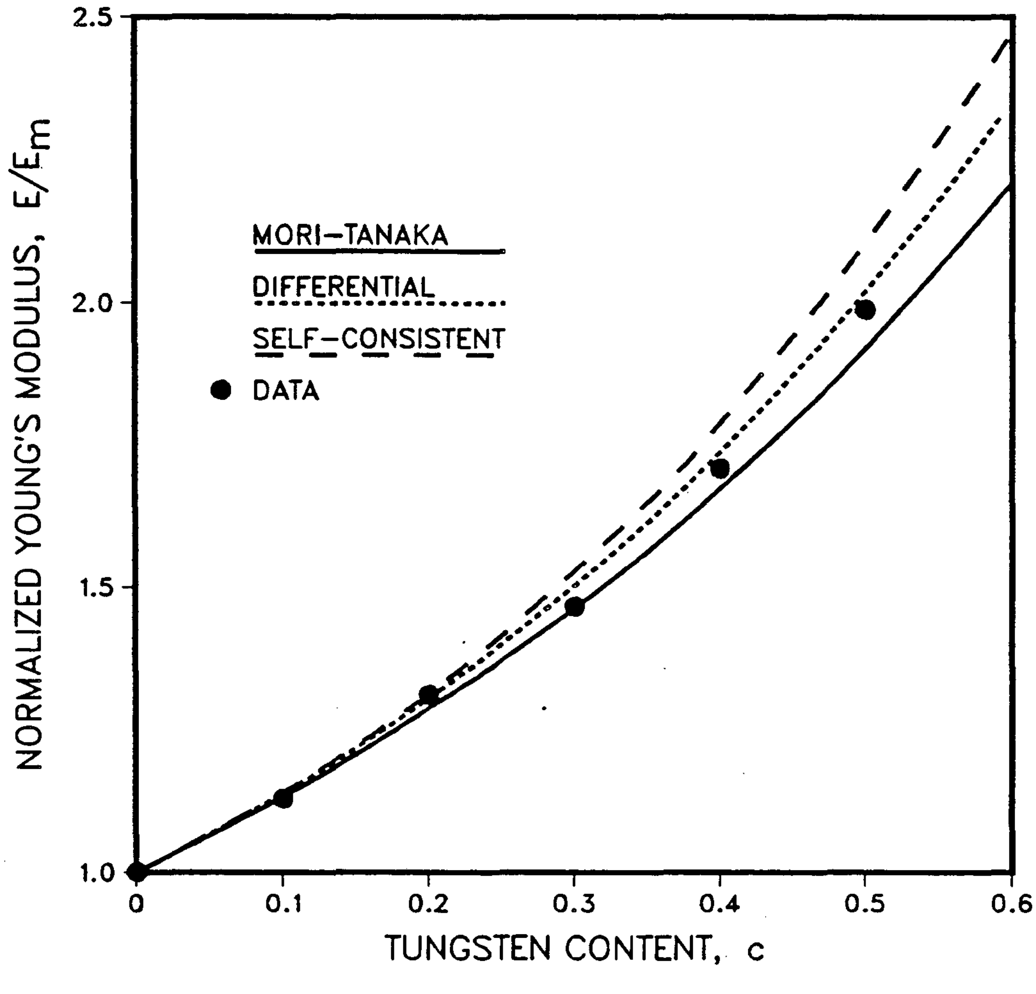

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Fig. 6. Young's modulus of a borosilicate glass containing spherical tungsten inclusions, compared with the predictions of the various theories. Data are from Hasselman and Fulrath (1965); elastic moduli of the two phases are listed in text.

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