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A Theoretical Method for Determination of Effective Elastic Constants of Isotropic Composites

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Abstract: A new self-consistent scheme is presented for determination of effective elastic constants of a composite having randomly oriented and randomly distributed spheroidal inclusions. In general, such a composite may be regarded as an isotropic and homogeneous material from the macroscopic point of view. Regarding the macroscopically homogeneous composite as a matrix and introducing additional inclusions of a sufficiently small volume fraction into the matrix, the effective elastic constants of the new composite are calculated by the use of a dilute theory. The new composite is also macroscopically isotropic and homogeneous. Carrying out the above procedure successively, we, thus, obtain the effective elastic constants of a composite with an arbitrary amount of volume fraction of inclusions. The numerical calculation confirms the validity of our method for all volume fractions of inclusions: The computed constants lie between the upper and the lower bounds of Hashin and Shtrikman, agree with the exact solution in the so-called Hill's case, and show reasonable asymptotic behaviour as the volume fraction of inclusions tends to unity.

1. Introduction

The theoretical determination of effective elastic property of composite material is still an up-to-date problem in the field of material science. Recently, this problem has become of practical importance also in the field of solid earth science. An example is found in studies on the physical and chemical feature of the low velocity zone of seismic waves in the upper mantle. The low velocity zone is explained presumably by the effective property of material composed of solid and melting phases. Another example arises from the observation of anomalous changes in seismic wave velocities prior to the occurrence of a large earthquake. The dilatancy is well-known phenomenon in the study of rock-fracture mechanics. When the deviatric stress applied to a rock sample becomes sufficiently large, the volumetric strain increases inelastically with an increase in the applied stress. This inelastic increase in volumetric strain is called dilatancy and considered to result from generation of micro-cracks in the sample. An increase in number of micro-cracks causes a decrease in velocity of elastic waves. Thus, the quantitative analysis of velocity changes observed in the source region of a large earthquake should be a powerful means not only to understand the physical process of earthquake occurrence but also to elucidate other possible premonitory phenomena.

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In the geophysical problems described above, the composite to be modeled for actual materials in the earth consists of phases whose elastic properties generally differ greatly from each other. Although a large number of studies have been made so far on the theoretical determination of effective elastic constants of composite materials, the existing theoretical methods do not seem accurate enough in the case of a large volume fraction of inclusions whose elastic property is of a great contrast to that of the matrix. The present paper aims to propose a new self-consistent scheme which will be abridged as NSC. The scheme will be applied to a composite having randonly oriented spheroidal inclusions whose spatial distribution is also random. The numerical results by our method will be presented to show its validity for all volume fractions of inclusions.

2. Critical Review of Existing Methods

A number of studies have been made on the theoretical determination of effective elastic constants of composite material. The problem under consideration is reduced essentially to the boundary value problem of the first kind or to that of the second kind, that is, the problem to find the stress field under the displacement condition given at the outer boundary of the composite or the strain field under the stress condition. The theoretical studies published so far may be classified into two groups. One intends to develop the so-called bounding method which gives us the upper and the lower bounds of effective elastic constants without any assumption for the geometry of its constitutents. The other is devoted to solve the boundary value problems for a composite containing inclusions of given geometry. The methods in this group result in the effective constants dependent on the shape of inclusions, and will be called the shape-dependent methods, hereafter.

(1) Bounding methods (shape-independent methods)

The Voigt and Reuss averages are well-known as classical theories for bounding methods. In order to derive his average, Voigt (1928) assumed that the strain field within a composite was approximated by a uniform strain field with the same magnitude as that of the applied strain to the composite. On the other hand, Reuss (1929) considered that the stress field in a composite was expressed approximately by a uniform stress with the same magnitude as that of the applied stress. Hashin and Shtrikman (1963) later showed that the Voigt and Reuss averages can be regarded as the upper and the lower bounds of effective elastic constants of a composite material. Hashin and Shtrikman (1962, 1963) greatly improved the bounding theory to derive the so-called HS bounds of effective elastic constants of multi-phase materials from their own variational principle. The difference between their upper and lower bounds was found to be sufficiently small for the composites consisting of solid phases whose elastic properties do not differ so much from one another. The HS bounding method is practically highly useful to determine the effective elastic constants of such composites as above. The method, however, loses its practical usefulness for the

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composite having void or liquid inclusions, because the lower bounds of rigidity and/or incompressibility become zero even for a very small volume fraction of inclusions.

There is a special cases where the rigidities of all constituent phases are the same (Hill's case). Hill (1963) found the exact solution for the effective elastic constants in this case. The effective rigidity is identical to that of constitutent phases and the effective incompressibility is uniquely determined independently of the shape of inclusions. The upper and the lower bounds of Hashin and Shtrikman for incompressibility are confirmed to degenerate to an identical value, which is consistent with the Hill's exact solution.

(2) Shape-dependent methods

Generally speaking, the shape-dependent theories are based on the solution of the boundary value problem of the first or the second kind for a composite material having inclusions of specified geometry. When the volume fraction of inclusions is so small that the interactions among inclusions may be negligible (dilute case), the well-known solution of the problem for a single inclusion is directly applicable to the determination of effective elastic constants. The shape-dependent methods in dilute case may thus be considered to have been well established. In non-dilute case in which the volume fraction of inclusions is not small enough, the solution for a single inclusion cannot be used without any modification. In conventional shape-dependent theories, therefore, the interaction effect among inclusions is taken into account in some approximate way.

There is another approach to derive a shape-dependent method, which makes use of the dynamic theory of wave scattering due to an inclusion of specific geometry. However, the situation about interaction effect is similar to the static treatment. The single scattering theory, which corresponds to the dilute case, is well established, while the multiple scattering theory has not yet been available.

(i) Methods for dilute concentration

Eshelby (1957) has presented the solution of strain field in an ellipsoidal inclusion in an infinite material which is strained uniformly at infinity, where both the two materials are assumed to be isotropic and homogeneous. He suggested a method for the determination of effective elastic constants by using his solution. One of the formulae derived from his method was obtained under the condition of fixed surface displacement, and the other under the condition of fixed surface traction. The formulae obtained from the two different boundary conditions are agree with each other to the first order term of volume fraction. This means that the method is valid for the case of dilute concentration of inclusions. In the Hill's case, the effective incompressibility determined by the methods is independent of the inclusion shape, but is not identical with the exact solution except for an infinitensimally small volume fraction, while the effective rigidity coincides with those of constitutent phases.

The approach using the theory of scattering waves was adopted by Mal and Knopoff (1967), Garbin and Knopoff (1973, 1975a, b), Kuster and Toksöz (1974a), *etc.* Their results are considered to be valid for a small volume fraction of inclusions, since

their methods are based on the single scattering calculation. In spite of this fact, it is interesting to note that the result obtained by the method of Kuster and Toksöz (1974a) agrees with the exact solution in the Hill's case.

(ii) Methods for non-dilute concentration

Hill (1965) developed a method called the self-consistent scheme (SCS), which is considered to be applicable to a composite with a large volume fraction of inculsions. The method pressumes that the average strain and the average stress within one of inclusions can be evaluated by replacing the surroundings of the inclusion by an imaginary material whose elastic constants coincide with the effective elastic constants to be determined. Hill (1965) showed that his method yields the same results from the dual approach; from the displacement condition and from the stress one.

The result of SCS in the Hill's case is known to agree with the exact solution. Moreover, the SCS results in general lie between the upper and the lower bounds of Hashin-Shtrikman for all values of volume fraction. Walpole (1969) obtained analytical expressions for effective elastic constants in the case of disc-shape inclusions, where the disc-shape means the oblate spheroid with a sufficiently small aspect ratio. His results are found to agree with the upper or the lower bounds of Hashin-Strikmann, according that the elastic constants of inclusions are larger or smaller than those of the matrix.

Budiansky (1965) has derived the same SCS expressions as Hill's (1965) in the case of spherical inclusions through a slightly different procedure. Using a method similar to that of Budiansky (1965), Wu (1966) calculated the effective elastic constants in the case of disc-shape and needle-shape inclusions, where the needle-shape means the prolate spheroid with sufficiently large aspect ratio. Watt *et al.* (1976) confirmed that the results by Wu (1966) are equivalent to those by Walpole (1969) in the case of discshape inclusions. Generally speaking, the numerical computation of successive approximation is required for SCS to find the effective elastic constants. O'Connell and Budiansky (1974) have presented analytical formulae for approximate solutions of th SCS equations in the case of void or liquid disc-shape inclusions.

Hill (1965) pointed out that the SCS equations in the case of void spherical inclusions have a positive root of effective elastic constant only when the volume fraction is less than 1/2, and stated that the results for large volume fractions are unreliable in such an extreme case as void inclusions. Bruner (1976) also has claimed that the SCS results in the case of void inclusions are physically unreasonable. Comparing the results of SCS with those obtained by scattering calculation to the second order, Chatterjee *et al.* (1978) concluded that the SCS results are not to be trusted beyond the first order in volume fraction of inclusions.

The numerical computation of higher order scattering is almost impossible at present, and has been attacked only in some special cases. For instance, Chatterjee *et al.* (1978) computed the scattering waves to the second order in the cases of void and rigid spherical inclusions. Computing SH-waves to the sixth order scattering, Varadan *et al.* (1978) determined only one of five effective moduli of a composite in the

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case where the inclusions of elliptic cylinder were oriented in the same direction.

Any approximation method proposed for effective elastic constants of a composite in non-dilute cases should be tested by the following criteria: The effective moduli determined by the method should (a) lie between the upper and the lower bounds of Hashin-Shtrikman for all volume fractions of inclusions, (b) show the correct asymptotic behaviour at sufficiently small volume fractions, (c) exhibit the physically reasonable asymptotic behaviour as the volume fraction tends to unity, (d) reduce to the exact solution of Hill (1963) in the Hill's case, and (e) satisfy the dual approach requirement, that is, the same result should be obtained under the fixed displacement condition and from the fixed stress condition on the outer boundary of a composite. Among these criteria, (a), (b), and (d) have been proposed by Watt *et al.* (1976).

3. A New Self-Consistent Scheme

The purpose of this section is to present a new method for calculation of effective elastic constants of a composite material which is macroscopically homogeneous. For simplicity, we treat here a two-phase composite having a single type of inclusions whose shapes and elastic properties are the same, though our method is easily generalized to the case of multi-phase composite. Further, the random orientations of spheroidal inclusions are assumed to make use of existing theory. That is to say, the composite concerned is macroscopically isotropic.

Consider a composite of unit volume which contains a finite volume fraction, v_k , of inclusions. Let us assume that effective elastic constants C_k can be defined for this composite and estimated by some means. In most cases, these effective constants are physically meaningful and practically useful, when the composite can be regarded as a macroscopically homogeneous material. For instance, let us consider the problem of elastic wave propagation. In this case, the wave length should be long enough compared with the size of inclusion, and, further, the spatial distribution of inclusions should be uniform in the composite. In logical consequence of this consideration, we may regard the microscopically heterogeneous composite as a macroscopicly homogeneous matrix with elastic constants C_k . Let us introduce an additional amount, dv_{k+1} , of inclusions into the macroscopically homogeneous matrix. If dv_{k+1} is sufficiently small compared with the volume of the matrix, the dilute theory is applicable to determine with a sufficient accuracy the effective elastic constants of the composite with volume fraction dv_{k+1} of inclusions.

Starting from a truly homogeneous matrix without any inclusion, we may repeatedly introduce an infinitesimally small volume fraction of inclusions into the macroscopically homogeneous matrix to give a composite with a finite value of volume fraction. At each successive iteration we may use the dilute method, and finally find the effective elastic constants of the composite having an arbitrary amount of inclusions. This is the basic concept of our method, which will be called new self-consistent scheme (NSC).

As explained above, a composite having a volume fraction v_k of inclusions is

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regarded as a macroscopically homogeneous matrix with elastic constants C_k . The relation between average stresses p and average strains e in the matrix is given by

$$\boldsymbol{p} = \boldsymbol{C}_k \boldsymbol{e} , \qquad (1)$$

and the strain energy E_k is expressed by

$$2E_k = (\boldsymbol{e} \cdot \boldsymbol{C}_k \boldsymbol{e}) = (\boldsymbol{p} \cdot \boldsymbol{C}_k^{-1} \boldsymbol{p}).$$
⁽²⁾

To introduce an additional small amount dv_{k+1} of inclusions into the matrix, we have to replace the volume dv_{k+1} of the matrix by the additional inclusions. In doing this, the given displacements or the given tractions at the outer surface of the matrix should be kept constant. The stress free strains e^{T}_{k} , defined by Eshelby (1957), due to insertion of the inclusions is expressed by

$$e^T{}_k = T_k e \,, \tag{3}$$

where T_k is a constant determined for given elastic properties of the matrix and inclusions of a specified shape. Following the theory of dilute concentration, the strain energy, E_{k+1} , in the composite after the introduction of additional inclusions is obtained as

$$2E_{k+1} = (\boldsymbol{e} \cdot \boldsymbol{C}_k \boldsymbol{e}) - d\boldsymbol{v}_{k+1} (\boldsymbol{e} \cdot \boldsymbol{C}_k \boldsymbol{T}_k \boldsymbol{e}), \qquad (4a)$$

for the fixed surface displacements, and

$$2E_{k+1} = (\boldsymbol{p} \cdot \boldsymbol{C}_k^{-1} \boldsymbol{p}) + dv_{k+1} (\boldsymbol{p} \cdot \boldsymbol{T}_k \boldsymbol{C}_k^{-1} \boldsymbol{p}), \qquad (4b)$$

for the fixed surface tractions. Writing E_{k+1} in terms of the effective elastic constants, C_{k+1} , of the new composite, we have

$$(\mathbf{e} \cdot \mathbf{C}_{k+1}\mathbf{e}) = (\mathbf{e} \cdot \mathbf{C}_k\mathbf{e}) - dv_{k+1}(\mathbf{e} \cdot \mathbf{C}_k\mathbf{T}_k\mathbf{e}), \qquad (5a)$$

or

$$(\boldsymbol{p} \cdot \boldsymbol{C}_{k+1} \boldsymbol{p}) = (\boldsymbol{p} \cdot \boldsymbol{C}_{k}^{-1} \boldsymbol{p}) + dv_{k}^{-1} (\boldsymbol{p} \cdot \boldsymbol{T}_{k} \boldsymbol{C}_{k}^{-1} \boldsymbol{p}), \qquad (5b)$$

from (4a) or from (4b).

Since the orientations of the newly introduced inclusions are also random, we obtain the following expressions from (5a) for the effective incompressibility K_{k+1} and for the effective rigidity μ_{k+1} of the new composite as

$$\frac{K_{k+1}}{K_k} = 1 - dv_{k+1} A(K_k, \mu_k, K^{(1)}, \mu^{(1)}, \alpha^{(1)}),$$

$$\frac{\mu_{k+1}}{\mu_k} = 1 - dv_{k+1} B(K_k, \mu_k, K^{(1)}, \mu^{(1)}, \alpha^{(1)}).$$
(6a)

From (5b)

$$\frac{K_k}{K_{k+1}} = 1 + dv_{k+1} A(K_k, \mu_k, K^{(1)}, \mu^{(1)}, \alpha^{(1)}),$$

$$\frac{\mu_k}{\mu_{k+1}} = 1 + dv_{k+1} B(K_k, \mu_k, K^{(1)}, \mu^{(1)}, \alpha^{(1)}).$$
(6b)

Here A and B are coefficients determined by K_k , μ_k of the matrix and $K^{(1)}$, $\mu^{(1)}$, and

 $\alpha^{(1)}$ of inclusions. The aspect ratio α is defined by $\alpha = c/a$, where *a* denotes the half length of axes of circular cross-section and *c* does that of the other axis of spheroid.

It should be noted that the total volume fraction after introduction of inclusions is not equal to $v_k + dv_{k+1}$. Pre-existing inclusions are randomly distributed in the matrix and the introduction of the additional inclusions into the matrix is done also randomly. Some of pre-existing inclusions are statistically expected to be overlapped by the additional inclusions. The overlapped part of the pre-existing inclusions must be replaced by the new inclusions or, in other expression, the overlapped part of the pre-existing inclusions. In this sense the overlapped part will be provisionally called the removed part in this paper. It is noted that the whole of a pre-existing inclusion is not necessarily overlapped by a new one. It is more probable that only a part of a pre-existing inclusion is overlapped and removed from the composite matrix. The removed part does not cause the real increase of volume fraction and should be subtracted from $v_k + dv_{k+1}$. The volume fraction of removed part is expected to be $v_k dv_{k+1}$. Therefore the real volume fraction after the introduction of new inclusions should be expressed as

$$v_{k+1} = v_k + dv_{k+1}(1 - v_k) . (7)$$

On the other hand, the quantity $v_k + dv_{k+1}$ will be called the nominal volume fraction, V_{k+1} , hereafter. As stated before the effective elastic constants of a composite with a given volume fraction of inclusions can be calculated by the successive use of (6a) or (6b) and (7) from k=0, where $v_0=0$.

Let us consider the limiting value of volume fraction as dv_k tends to zero. If we introduce the same amount, dv, of inclusions at every iteration, the real volume fraction v_n and the nominal volume fraction V_n at the *n*-th iteration are written by

$$v_n = 1 - \left(1 - \frac{V_n}{n}\right)^n,$$

$$V_n = n dv,$$
(8a)

The volume fraction of removed part, v_{rn} , which is the total volume fraction of pre-existing inclusions replaced with the newly introduced inclusions at every iteration, is expressed by

$$v_{n} = V_n - v_n \tag{8b}$$

Consider the case where the real volume fraction v is attained by successive introduction of inclusions of an infinitesimally small volume fraction. The nominal volume fraction V is obtained from (8a) as

$$V \equiv \lim_{dv \to 0} V_n = -\ln\left(1 - v\right). \tag{9}$$

The volume fraction of removed part v_r is written from (8b) as

$$v_r \equiv \lim_{dv \to 0} v_{rn} = -\ln(1-v) - v.$$
 (10)

It is shown from (9) and (10) that V and v_r are finite values, except for the special case of v=1.

We cannot express analytically the limiting values of effective elastic constants calculated by the successive use of (6a) or (6b). Their convergency, however, is apparent for a finite value of V, as far as the coefficients A and B in (6a) or in (6b) have finite values.

Further if dv is taken to be small enough, practically equal effective elastic constants are calculated either form (6a) or from (6b) at each iteration. We can have the result as accurate as we desire by taking a smaller value of dv. We, therefore, obtain the same result by the successive use of either expression. Our method of NSC is thus considered to satisfy the dual approach requirement stated in Section 2.

4. Numerical Results for Two-Phase Composites

The effective elastic constants are numerically calculated by the new selfconsistent method (NSC) for various two-phase materials. In order to examine the validity of the method, the results are compared with those by other methods such as SCS by Hill (1965), Wu (1966), Walpole (1969) and O'Connell and Budiansky (1974), KT by Kuster and Toksöz (1974a), DSP by the expressions in the case of dilute concentration based on the displacement condition, and STR by the expressions for the dilute case based on the stress condition, as well as the upper (HS⁺) and the lower (HS⁻) bounds of Hashin and Shtrikman (1963).

(1) Hill's case

The effective elastic constants obtained by various methods in Hill's case are illustrated in Fig. 1. As is well known, the upper bound, HS⁺, for the effective incompressibility coincides with the lower bound, HS⁻, and is equal to the Hill's exact solution (Hill, 1963). Although the effective incompressibilities by DSP and STR are independent of inclusion shape, they differ significantly from the exact solution. It is found, however, that the present method (NSC) based on DSP or STR expressions for dilute concentration gives the same result as the exact solution. The same results can be obtained also by SCS and KT independently of inclusion shape. The effective rigidity obtained by every method is identical with that of the matrix and inclusions.

(2) Spherical inclusions

The results in the cases of void and liquid spherical inclusions are shown in Figs. 2 and 3, respectively. The lower bounds HS⁻ for incompressibility and for rigidity are equal to zero in the case of void inclusions for all values of volume fraction, and those for rigidity are also equal to zero in the case of liquid inclusions. These are omitted from the figures. The NSC results lie between the upper and the lower bounds of HS for any value of volume fraction. The effective elastic constants by NSC are found to tend to those of inclusions as the volume fraction approaches to unity. The results of SCS are fairly close to those of DSP in both the cases of void and liquid inclusions. In the results of SCS the effective incompressibility and rigidity vanish at the volume fraction of 0.5 in the case of void inclusions, and the effective rigidity does at a volume fraction near 0.6 in the case of liquid inclusions. The result of KT coincide with those of $\rm HS^+$ in both the cases.

The upper bound HS⁺ for incompressibility is known to correspond to the effective incompressibility of a spherical material having a concentric spherical inclusions. Therefore, it is reasonable to expect that the NSC results should be less than HS⁺, since a uniform distribution of small inclusions is presumed for NSC. The figures show



- Fig. 1. Effective incompressibility K calculated by various methods in Hill's case ($\mu^1 = \mu_0$). K, μ , and ν denote incompressibility, rigidity, and Poisson's ratio, respectively. The quantities with subscript 0 and superscript 1 indicate those of matrix and of inclusion, respectively.
 - NSC; the new self-consistent method in our study,
 - $\mathrm{HS}^{\pm};$ the upper and the lower bounds of Hashin and Shtrikman,
 - DSP; the dilute method from the displacement condition,
 - STR; the dilute method from the stress condition,
 - KT; the method of Kuster and Toksöz,
 - SCS; the method of self-consistent scheme.
 - $\mathrm{HS}^{\pm},$ NSC, KT, and SCS are all equal to the exact solution of Hill.



Fig. 2. Effective incompressibility \overline{K} and effective rigidity $\overline{\mu}$ in the case of void spherical inclusions. The notations are the same as those in Fig. 1.



Fig. 3. \overline{K}/K_0 and $\overline{\mu}/\mu_0$ in the case of liquid spherical inclusions.



Fig. 4. \bar{K}/K_0 and $\bar{\mu}/\mu_0$ in the case of disc-shape inclusions. Calculations are done by putting $\alpha = 10^{-3}$ except for SCS. SCS are calculated for inclusions with infinitesimally small aspect ratio and are practically equal to NSC and to HS⁻.

further that the STR results are the worst, because they are larger than those of HS⁺ for any value of volume fraction.

(3) Disc-shape inclusions

An oblate spheroidal inclusion with a sufficiently small aspect ratio is here called a disc-shape one for the sake of convenience. An example is shown in Fig. 4 for the case where inclusions have finite and small values of elastic constants. Calculations of NSC, KT, STR and DSP are performed in the case of $\alpha = 10^{-3}$. For SCS, the expressions presented by Walpole (1969) are used, which are established to be valid when the aspect ratio of inclusions is sufficiently small compared with the ratios of elastic constants of inclusions to those of the matrix, provided that the elastic constants of inclusions are less than those of matrix. The results of NSC are found to approach those of HS⁻ as the aspect ratio decreases, and they are, as shown in the figure, nearly equal to those of HS⁻ when $\alpha = 10^{-3}$. Walpole (1969) has proved analytically that the results of SCS are identical with those of HS⁻ in this case. Although KT is known to be valid only when the concentration of inclusions is dilute, it gives a better approximation to the results of NSC than the DSP. The results of STR are accidentally close to those of NSC. Fig. 5 illustrates the results of NSC in the case of void inclusions for some values of aspect ratio, where the crack density v_0 defined by $v_0 = v/\alpha$ is used instead of the volume fraction v. It is shown in the figure that the effective incompresibility and the effective rigidity, respectively, tend to certain values for a given crack density, as the aspect ratio decreases. This means that it is convenient to use crack density v_0 instead of v as a parameter expressing the concentration of inclusions in the case where the aspect ratio of void inclusions is sufficiently small. The results in this case are plotted against crack density in Fig. 6. The SCS results are computed by the use of the expressions presented by O'Connell and Budiansky (1974). The KT is found to give the values closest to those of NSC. It is noted that the SCS evaluates effective rigidity smaller than any other methods.

(4) Needle-shape inclusions

The effective elastic constants of NSC are easily confirmed to have some limiting values as the aspect ratio increases to infinity when the volume fraction of inclusions is kept constant. The shape of inclusion in this case is called needle shape. The results in the cases of void and liquid inclusions are shown in Figs. 7 and 8. The NSC results lie between the upper and the lower bounds of HS for all values of volume



Fig. 5. \bar{K}/K_0 and $\bar{\mu}/\mu_0$ of NSC versus crack density $v_0(v_0 = v/\alpha)$ in the case of void inclusions.



Fig. 6. \bar{K}/K_0 and $\bar{\mu}/\mu_0$ versus crack density in the case of void disc-shape inclusions. SCS are calculated from the formulae given by O'Connell and Budiansky (1974).



Fig. 7. \overline{K}/K_0 an $\overline{\mu}/\mu_0$ in the case of void needle-shape inclusions.



Fig. 8. \overline{K}/K_0 an $\overline{\mu}/\mu_0$ in the case of liquid needle-shape inclusions.

fraction and tend to the elastic constants of inclusions as the volume fraction approaches to unity. The SCS results calculated by the expressions presented by Watt *et al.* (1976) are very close to those of DSP.

5. Generalization to Multi-Phase Composites

Consider a composite material of unit volume which contains N kinds of inclusions. They are different in elastic properties and/or in shape of inclusions. The inclusions of each kind are oriented at random and distributed homogeneously. This composite material can also be regarded as an isotropic and homogeneous material in a macroscopic sense, its incompressibility and rigidity being denoted by K_k and μ_k , respectively. Let us introduce simultaneously small amounts $dv_{k+1}^{(i)}$ of all the kinds of inclusions into the composite matrix having effective constants of K_k and μ_k , where $dv_{k+1}^{(i)}$ means the volume fraction of the *i*-th kind inclusions at the (k+1)-th iteration. As far as the sum of $dv_{k+1}^{(i)}$ with respect to (*i*) is small enough, the effective elastic constants of the composite that contains the newly introduced inclusions can be evaluated by the use of the expressions valid for the dilute concentration of inclusions similarly to the case of two-phase materials as

$$\frac{K_{k+1}}{K_k} = 1 - \sum_{i=1}^{N} dv_{k+1}{}^{(i)} A(K_k, \mu_k, K^{(i)}, \mu^{(i)}, \alpha^{(i)}),
\frac{\mu_{k+1}}{\mu_k} = 1 - \sum_{i=1}^{N} dv_{k+1}{}^{(i)} B(K_k, \mu_k, K^{(i)}, \mu^{(i)}, \alpha^{(i)}),$$
(11a)

or

$$\frac{K_{k}}{K_{k+1}} = 1 + \sum_{i=1}^{N} dv_{k+1}{}^{(i)} A(K_{k}, \mu_{k}, K^{(i)}, \mu^{(i)}, \alpha^{(i)}),
\frac{\mu_{k}}{\mu_{k+1}} = 1 + \sum_{i=1}^{N} dv_{k+1}{}^{(i)} B(K_{k}, \mu_{k}, K^{(i)}, \mu^{(i)}, \alpha^{(i)}),$$
(11b)

$$dv_{k+1} = \sum_{i=1}^{N} dv_{k+1}^{(i)} , \qquad (12)$$

where the definition of quantities is the same as that in previous sections taking that the quantities with superscript (i) denote those of the inclusions of the *i*-th kind. The total volume, v_{k+1} , of all inclusions is expressed by (7). For the real volume fractions of inclusions of the *i*-th kind, we have

$$v_{k+1}^{(i)} = v_k^{(i)} - v_k^{(i)} dv_{k+1} + dv_{k+1}^{(i)} .$$
(13)

according to the same consideration as explained in the two-phase case. We can evaluate the effective elastic constants of the composite containing N-kinds of inclusions by the successive use of (11a), or (11b), and (12) for arbitrary values of $v_n^{(i)}$.

In the above procedure all kinds of inclusions were introduced into the matrix simultaneously. It is, of course, possible to introduce one kind of inclusions after another successively. The above expressions are valid also in this case. The manner of introduction has an effect on the removed volume fraction of inclusions of a kind and, consequently, on the estimated effective elastic constants. To examine this in detail, we consider the case of $dv_k = dv = const$. at all iterative steps. For given values of $dv_k^{(i)}$ at the k-th step of iteration, we further define

$$\left. \begin{array}{c} r_{k}^{(i)} = dv_{k}^{(i)}/dv \\ \sum_{i=1}^{N} r_{k}^{(i)} = 1 \,. \end{array} \right\}$$

$$(14)$$

Let us consider first the case of simultaneous introduction. In this case $r_k^{(i)} = r^{(i)} = const.$ at all steps. The real volume fraction $v_n^{(i)}$ of inclusions of the *i*-th kind at the *n*-th interation is given by $v_n^{(i)} = r^{(i)}v_n$, where v_n is the total volume fractions of all the inclusions as obtained by (8). The nominal volume fraction $V_n^{(i)}$ of the *i*-th kind inclusions is evidently written as $V_n^{(i)} = r^{(i)}V_n$, where $V_n = ndv$. Therefore, the difference between nominal volume fraction and real one is equal to the volume fraction $v_{rn}^{(i)}$ of removed part of the *i*-th kind inclusions. Similarly to (9) and (10) in the case of two-phase composite, we have

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$$\begin{cases} V^{(i)} = \lim_{dv \to 0} V_n^{(i)} = -r^{(i)} \ln (1-v) , \\ v_r^{(i)} = \lim_{dv \to 0} v_{rn}^{(i)} = r^{(i)} \{ -\ln (1-v) - v \} , \end{cases}$$
(15)

where v is given.

Next, we consider the case of one-by-one introduction. In this case, we successively introduce inclusions of a kind chosen arbitrarily from many kinds of inclusions. After introduction of the first kind is completed, another kind is successively inserted in the matrix. The same procedure is repeated N times, where N is the number of kinds of inclusions. Following the previous formulation, we have

where n_i is the number of iterations necessary to complete introduction of inclusions to the *i*-th kind. For the sake of simplicity, consider the case of three-phase material, that is, N=2. Putting $n_1=m$ and $n_2=n>m$, (8) gives

$$v_m^{(1)} = 1 - (1 - dv)^m$$
,
 $v_m^{(2)} = 0$,

because the composite is a two-phase material up to the m-th step of iteration. At the n-th iteration, we have

$$v_n^{(1)} = v_m^{(1)} (1 - dv)^{n-m} = (1 - dv)^{n-m} - (1 - dv)^n,$$

$$v_n^{(2)} = 1 - (1 - dv)^{n-m}.$$
(17)

These are the real volume fraction of the first and second kind inclusions. The real total volume fraction of all inclusions, v_n , is written by

$$v_n = 1 - (1 - dv)^n \,. \tag{18}$$

This relation (18) should always be satisfied irrespective of the manner of inclusion introduction. The nominal volume fractions of the first and the second kinds are

$$\begin{cases} V_{n}^{(1)} = m dv, \\ V_{n}^{(2)} = (n - m) dv. \end{cases}$$
(19)

Assuming that prescribed values of $v^{(1)}$ and $v^{(1)}$ are given to $v_n^{(1)}$ and $v_n^{(2)}$, respectively, we have

$$V^{(1)} = \lim_{dv \to 0} V_n^{(1)} = -\ln\left(1 - \frac{v^{(1)}}{1 - v^{(2)}}\right),$$

$$v_r^{(1)} = \lim_{dv \to 0} v_{rn}^{(1)} = \lim_{dv \to 0} (V_n^{(1)} - v^{(1)}) = -\ln\left(1 - \frac{v^{(1)}}{1 - v^{(2)}}\right) - v^{(1)},$$

$$(20)$$

$$V^{(2)} = \lim_{dv \to 0} V_n^{(2)} = -\ln(1 - v^{(2)}) ,$$

$$v_r^{(2)} = \lim_{dv \to 0} v_{rn}^{(2)} = \lim_{dv \to 0} (V_n^{(2)} - v^{(2)}) = -\ln(1 - v^{(2)}) - v^{(2)} .$$

$$(21)$$

Even in the case of $v^{(1)}=v^{(2)}$, (20) and (21) indicate that the nominal volume fraction of the first kind inclusions is larger than that of the second kind. The same is true also for the volume fraction of removed part of inclusions. Returning to the case of simultaneous introduction, we put N=2, $v/2=v^{(1)}=v^{(2)}$, and $r^{(1)}=r^{(2)}=1/2$ in (15). In this case we have the same nominal volume fraction for the first and second kinds, and its value lies between $V^{(1)}$ and $V^{(2)}$ in the case of one-by-one introduction. This suggests that different values of effective elastic constants are possibly estimated by different ways of inclusion introduction even for the same real volume fractions of constitutent phases.

In order to show this numerically, we take an extreme case of three-phase composite, in which the first kind of inclusions is very soft and the second very hard compared with the matrix, as listed in Table 1 (a). The numerical results of effective elastic constants are given in Table 1(b). Cases I and II are both the one-by-one introduction. In case I, the soft material is first introduced into the matrix and the hard one is done next. Case II is the reversed case. The calculated values of elastic constants are smaller in case I than those in case II. Case III is the simultaneous introduction and its results fall between cases I and II. The phase that was introduced

Table 1 Calculated Results for Three-Phase Composite

(a) Parameters Used for Calculation

	K	μ	æ	υ
Matrix	1.0	0.6		0.8
Phase 1	0.1	0.06	0.5	0.1
Phase 2	10.0	6.0	0.5	0.1

K; Incompressibility. μ ; Rigidity.

 $\boldsymbol{\alpha}$; Aspect ratio. v; Volume fraction.

(b)	Results	of	\overline{K}/K_0	and	Tu/no

Case	\overline{K}/K_0	$\overline{\mu}/\mu_0$
I	0,953	0.993
II	0.977	1.018
III	0.965	1.005

 \overline{K} , $\overline{\mu}$; Effective incompressibility and rigidity of composite. K_0 , μ_0 ; Incompressibility and rigidity of matrix.

- I; The case in which Phase 1 is introduced first and Phase 2 next.
- II; Reversed case of I.
- III; The case in which Phases 1 and 2 are simultaneously introduced.

and

Table 2 Calculated Results in the Hill's Case of Three-Phase Composite

	K	μ	v
Matrix	1.0	0.09375	0.8
Phase 1	0.1	0.09375	0.1
2	10.0	0.09375	0.1

(a) Parameters Used for Calculations

K, μ , v; The same as those in Table 1 (a).

(b) Results of K/K_0

Case	\overline{K}/K
Exact Sol.	0.7331
I	0.7336
II	0.7335
III	0.7335

 \overline{K} , K_0 , I, II, III; The same as those in Table 1 (b).

first has a stronger effect on the calculated overall property of the composite. This is due to the difference in nominal volume fraction of the two phases.

A large nominal volume fraction implies a relatively large number of mutually connected inclusions of the same kind. Some of inclusions of the kind introduced first are subjected to partial replacement by inclusions of the second kind and change their shapes into different ones. On the other hand, the change in shape due to the other phase does not occur for inclusions introduced second. Therefore, the minute structure of a composite depends necessarily on the detailed process in producing it in the case of multi-phase material, but not in the case of two-phase material. If we may assume, however, that the distribution of inclusions of all kinds are completely random in a composite, the use of simultaneous introduction is recommended.

To examine the above interpretation we present numerical results for a three-phase material in Hill's case, for which we have exact solutions of effective elastic constants independently of inclusion shape. Table 2 exhibits the computed effective incompressibility as well as constants of the three phases in this particular example, where the cases of I, II and III are the same as in Table 1(a). The difference in value of the three cases are smaller than computational errors. The results are independent of the manner of inclusion introduction, as was expected previously. The difference in effective incompressibility between the exact solution and three cases are slightly bigger than the differences among the three cases. However, the differences are found to be within numerical errors arising from our computation in the single precision mode.

6. Comparison of the Present Results with Experimental Data

The velocities and the attenuation of ultra-sonic waves were measured by Kuster

and Toksöz (1974b) in suspensions of solid spherical particles in liquid matrices. We compare the velocities calculated by NSC and SCS with their experimental data. This case of liquid matrix with solid inclusions is an exceptional case, where the effective rigidity determined by the dilute method of DSP, STR, or KT is always zero independently of the rigidity of inclusions. If we use the results by DSP or by STR as the basis of calculation, therefore, NSC gives also vanishing effective rigidity irrespectively of the volume fraction. Giving a sufficiently small value to the rigidity of the matrix, however, we can calculate the asymptotic behaviour as the volume fraction aproaches to unity. Fig. 9 illustrates such an asymptotic behaviour of effective rigidity determined by NSC, the results of effective incompressibility being shown as well. The effective incompressibility by NSC, of course, approaches to that obtained by putting zero for the rigidity of matrix, as the rigidity tends to zero.



Fig. 9. \overline{K}/K_0 and $\overline{\mu}/\mu_0$ of NSC in the case of solid spherical inclusions.



Fig. 10. Comparison of the theoretical velocities of P-waves with the experimental ones (solid circles) by Kuster and Toksöz (1974b) in the case of solid spherical inclusions in a liquid matrix.

- (a); The case of water and polystyrene.
- (b); The case of ATB (acetylene tetrabromide-benzine mixture) and glass.
- (c); The case of oil and polystyrene.
- \overline{V}_p and V_{po} being P-wave velocities of composite and matrix.

	Case (a)	Case (b)	Case (c)
Density Ratio of Inclusion to Matrix	1.05	1.02	1.19
V_{p} Ratio of Inclusion to Matrix	1.60	6.62	1.60

Table 3 Parameters Used for Calculations in the Cases of (a), (b), and (c) of Fig. 10

The SCS expressions by Watt et al. (1976) and KT can be used in the present case of spherical solid inclusions in a liquid matrix, where the rigidity of matrix is taken to be zero. It is remarked here that the KT results agree exactly with the lower bounds of HS. We calculate the effective velocities of compressional waves by NSC, SCS and KT, using the material constants presented by Kuster and Toksöz (1974b) as shown in Table 3. The results are illustrated in Figs. 10(a), (b) and (c) in comparison with the experimental results, where the velocities are normalized by those of matrices. The SCS results differ significantly from the experimental ones at volume fractions larger than 0.4. On the other hands, the NSC results, which are numerically almost equal to the KT results, agree well with the experimental data. The experimental results, however, are a little less than the NSC ones by 1 to 2% at larger volume fractions. This may be explained, at least in part, by the inelastic behaviour of the material, because the velocities of body waves are expected to decrease by 0.5% if the specific quality factor Q for inelastic attenuation is assumed to be 100 in the linear viscoelastic model of attenuation used by, for example, Kanamori and Anderson (1977).

7. Summary and Discussions

We presented in this study a new self-consistent scheme for determination of effective elastic constants of composite materials. In this scheme, the effective elastic constants even for materials with considerably high concentration of inclusions can be estimated by the successive use of the formulae for dilute concentration of inclusions. Any dilute theory can be used in this successive approximation. This scheme can be called self-consistent in the senses that the interaction energy of inclusions is evaluated in terms of the effective elastic constants and the average strain or the average stress over the composite, and also that the results by this method are independent of the adoption of the displacement or the stress condition given on the surface of the composite. The overall elastic moduli evaluated by the method were found to lie between the Hashin and Shtrikman bounds for all volume fractions, although the results of only several cases were presented in the figures. In particular, our method proved to be valid even in extreme cases of void and liquid inclusions with disc, sphere or needle shape. These are considered to be the cases where the effect of inclusions is prominent on the effective elastic property. It is concluded, therefore, that our method always satisfies the requirement of Hashin-Shtrikman bounds for any volume fraction of

inclusions irrespective of inclusion shape and elastic properties of matrix and inclusions. The effective elastic constants obtained by our method were found to tend asymptotically to the elastic constants of the inclusion material, as the volume fraction of inclusions approaches to unity. It was also shown that our results agree with the exact solution by Hill (1963) in the case where all the phases have the same rigidity. Thus, the results by our method completely satisfy all the requirements given in Section 2.

The effective elastic constants of a composite depend, in general, on the spatial distribution of inclusions in it. An interesting example is shown in Fig. 11 for a twophase composit having spherical inclusions. It is well known that the upper (lower) bound of Hashin and Shtrikman for effective incompressibility corresponds to that of spherical material having a concentric spherical inclusion, when the elastic constants of inclusion are smaller (larger) than those of matrix. Since our NSC method is based on the assumption of a random distribution of small inclusions, it is reasonable that our results in the case of spherical inclusions do not coincide with HS⁺(HS⁻).

SCS method for spherical inclusions gives an unreasonable result that the same effective elastic constants are estimated even when the hard material of matrix and soft one of inclusions are exchanged. They depend only on the volume fraction of a particular phase. Consider that the soft phase is now taken to be the matrix material. It is peculiar that the SCS gives the same estimates as in the previous case of soft inclusions under the assumption of spherical inclusions, because the shape of the soft matrix must be quite different from sphere. In other words, SCS assumes the shape of inclusions in one sense but is independent of the shape in another sense. As shown in Fig. 11, NSC predicts larger effective constants in the case of soft spherical inclusions compared with those in the case of hard ones. It may be concluded to be very natural.

Let us further examine the SCS results in the special case of a two-phase composite



Fig. 11. Comparison of the results calculated by HS[±], NSC, and SCS in the case of spherical inclusions. The upper curves of NSC indicate K and μ in the case of inclusions of soft material (B) in the matrix of hard material (A), and the lower curves do in the case of inclusions of A in the matrix of B. The SCS results in the two cases are identical with each other.

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having spherical void inclusions. As shown in Fig. 2, the SCS predicts that both the effective incompressibility and the effective rigidity vanish at volume fraction not less than 0.5. When spherical inclusions of an equal size are located at all grid points with spacing equal to the diameter of the spherical inclusion, the volume fraction of inclusions is about 0.524. In the closest packed case of spherical inclusions the volume fraction is about 0.74. Even in this cases the frame of solid matrix is still left, and some finite values for the effective elastic constants must be obtained, whereas SCS gives null values. This indicates that a particular arrengement of inclusions is required for the effective elastic constants to vanish at a volume fraction not less than 0.5. It was shown in Section 4 that the effective elastic constants by NSC continuously tend to the elastic constants of inclusions as the volume fraction of inclusions increases to unity.

The effective elastic constants by NSC were found to agree well with the experimental results by Kuster and Toksöz (1974b) in the case where solid spherical inclusions were suspended in a liquid matrix, as stated in Section 6. This is an envidence for the practical applicability of our method even to a high concentration of inclusions, while the SCS results disagree with the experiment at volume fractions greater than 0.4.

In the case of multi-phase composite, the results of our method depend on the manner of inclusion introduction, as described in Section 5. Let us consider the case of two different kinds of inclusions for a simple explanation. One of iterative processes in our method is that a kind (A) of inclusions is introduced first and another kind (B) of inclusions is introduced after the completion of insertion of A. This is called one-by-one introduction in this paper. The reverse process to introduce B first and later A is also possible. The disadvantage of our method is that the estimated effective elastic constants are different from each other even though the same volume fractions are assigned to respective kinds of inclusions. This disadvantage, however, can be avodied by adoption of simultaneous introduction process, which is to introduce small amounts of inclusions of both kinds simultaneously in each step of iteration. This simultaneous introduction assures us of unique determination of effective elastic property for a multi-phase composite.

Finally it is noted that the method by Kuster and Toksöz (1974a) for dilute concentration generally gives better estimates of the effective elastic constants than other dilute methods. Therefore, the use of the KT formulae is recommended at the basis of our method because it usually gives a faster convergency in the iteration process.

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