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POSTBUCKLING BEHAVIOR OF A PERIODICALLY LAMINATED MEDIUM IN COMPRESSION

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Abstract—A theory is constructed for geometrically nonlinear behavior of a periodically laminated medium for a class of planar deformation modes which are macroscopically homogeneous in the direction normal to the material interfaces. The theory is used to study microbuckling and initial postbuckling behavior of the medium under compression along the laminates. The results indicate that the bifurcation is of the stable symmetric type so that geometrical imperfections in the form of initial waviness of the reinforcements are not expected to reduce the microbuckling stress significantly.

INTRODUCTION

For a unidirectionally reinforced composite subjected to compression along the direction of the reinforcements, a mode of failure that is of some interest is microbuckling which occurs due to loss of uniqueness of the macroscopically homogeneous state of uniaxial compressive stress at sufficiently high values of the applied load. Although microbuckling can occur either in the extension mode or in the shear mode, where the appellation refers to the predominant state of incremental strain in the matrix material immediately after buckling, the shear mode of buckling is of greater relevance for the practically significant cases in which the volume fraction of the reinforcements is not too low.

In what follows we present an analysis of shear mode buckling as well as initial postbuckling behavior of a unidirectional composite in compression. Specifically, we consider a periodically laminated medium and assume that the constitutive equations of the two isotropic elastic constituents are linear relationships between Piola–Kirchoff stress of the second kind and the Lagrangian strain tensor, or, equivalently, that the respective strain energy functionals are quadratic in the strain components. Thus, for this particular choice of stress and strain measures, our theory contains only geometrical nonlinearity. Using this material model for the constituents we derive a nonlinear continuum model for the composite similar to the effective stiffness theory[1] for a restricted class of planar deformation modes and then present analyses of buckling and initial postbuckling behavior of the composite subjected to compression. Evidently, the object of initial postbuckling analysis is to determine, in the context of Koiter's theory of elastic stability[2], the influence of initial geometric imperfections on the magnitude of microbuckling stress. The initial postbuckling analysis also yields a quantitative estimate of reduction in the stiffness of the composite due to microbuckling.

The scope, emphasis and methodology of our study as outlined above are considerably different from those of previous micromechanics-based investigations (e.g. [3–9]) of the general problem of stability of a unidirectional composite in compression. Although in some of these studies even such complicating yet important factors as fiber debonding[7] and material nonlinearity have also been included, most are devoted to linear stability analysis and in all of them—except in the excellent treatment of buckling of imcompressible laminated media by Biot[3]—a beam on some type of elastic foundation has been employed as a model for composite behavior. In contrast to this type of material model, the so-called ideal fiber reinforced composites containing densely distributed inextensible fibers have been considered in the linear stability analysis by Kurashige[10] who allows the matrix to be compressible and in the postbuckling analysis due to Kao and Pipkin[11] wherein matrix incompressibility has been assumed. Our object in the sequel is to model the instability phenomenon on the basis of a continuum theory which takes into account the composite microstructure and is a more satisfactory description of material behavior than are the material models previously utilized.

FORMULATION

We consider a periodically laminated medium consisting of alternate layers of reinforcements and the matrix material, with perfect bonds assumed at the material interfaces. We also make the assumption that, with the geometry and coordinate system shown in Fig. 1(a), the composite is unbounded along the two coordinate directions x_2 and x_3. In the following development our interest is in the construction of a nonlinear theory of deformation in the planar modes of the type shown in Fig. 1(b) which are associated with shear mode buckling of the medium. These modes are homogeneous on the macroscale along the direction perpendicular to the laminates and consequently it is sufficient for our purposes to assume that the displacements are periodic along this direction, with length of periodicity equal to the unit cell dimension.

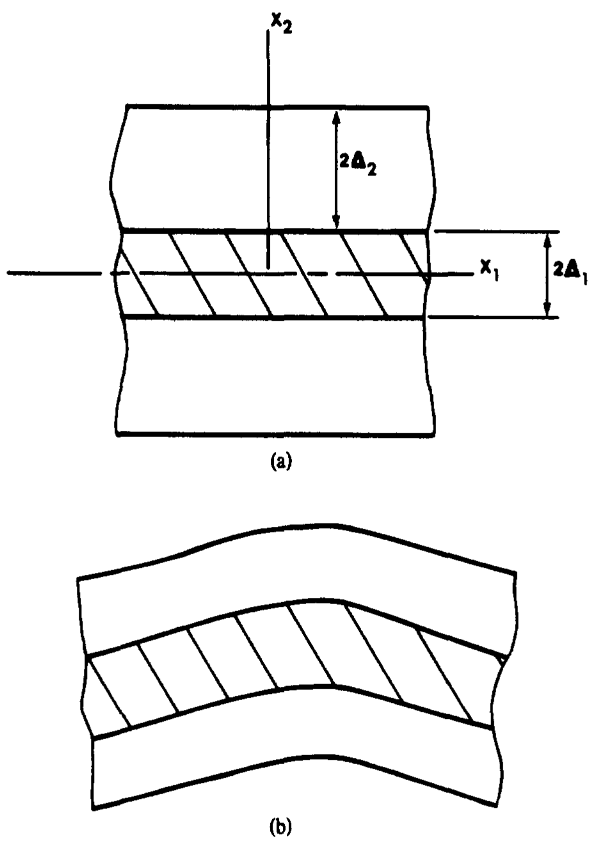
The model construction is based upon extremization of potential energy with respect to appropriate displacement fields whose variation in the transverse direction is assumed in such a form that it is possible to adquately represent the shear mode buckling pattern. We begin the analysis by noting that in plane strain the average strain energy density of the composite for periodic deformation is given by

$$V=\frac{1}{2\Delta}\left[\int_{\Omega^{(1)}} V_1\,\mathrm{d}x_2+\int_{\Omega^{(2)}} V_2\,\mathrm{d}x_2\right], \tag{1a}$$

with

$$V_\alpha=\frac{1}{2}[C_{11}^{(\alpha)}(e_{11}^2+e_{22}^2)+2C_{12}^{(\alpha)}e_{11}e_{22}+4C_{66}^{(\alpha)}e_{12}^2], \qquad 1b)$$

where $\Delta=\Delta_1+\Delta_2$ is one half of the length of the unit cell (see Fig. (1a)). Further, $\Omega^{(\alpha)}$ denotes



(b)

Fig. 1. (a) Geometry and coordinate system, (b) Shear mode buckling pattern.

the part of a typical unit cell occupied by the α constituent, with $\alpha = 1$ or 2 denoting the fiber and the matrix, respectively, and $C_{ij}^{(\alpha)}$ are appropriate functions of the Young's modulus and Poisson's ratio. The quantities e_{ij} in (1b) are the components of the Lagrangian strain tensor given by the usual strain displacement relations.

To compute the composite strain energy density we assume the variation of the displacement components within a unit cell to be of the form

$$u_1^{(\alpha)} = U(x_1) + \psi(x_1) y_2^{(\alpha)}/n_\alpha \tag{2a}$$

$$u_2^{(\alpha)} = W(x_1) + \phi(x_1) y_2^{(\alpha)}/n_\alpha \tag{2b}$$

where

$$y_2^{(1)} = x_2, \quad y_2^{(2)} = (\Delta - x_2); \quad -\Delta_1 \le x_2 \le \Delta_1 + 2\Delta_2, \tag{2c}$$

and n_α denotes the volume fraction of the α constituent. The assumed displacement profiles can be continued along the x_2 direction in such a manner as to yield kinematically admissible and periodic displacement fields with the length of periodicity being the unit cell dimension. Furthermore, these fields are consistent with the shear mode buckling pattern in which we are interested and they are simplest such profiles.

Another set of approximations that we introduce is the one corresponding to the usual small strains, moderate rotations approximation of the conventional nonlinear theories of plates and shells. Thus we assume that the x_1 dependent functions that occur in (2) satisfy

$$(U', \phi) = O(\epsilon^2); \quad (W', \psi) = O(\epsilon), \tag{3}$$

where $(\ )' \equiv \partial(\ )/\partial x_1$, and ϵ is a small parameter denoting the ratio of the unit cell half width, Δ, to a macrodimension.

On substitution of the assumed displacement profile in the strain-displacement relations we obtain

$$e_{11}^{(\alpha)} = U' + \frac{y^{(\alpha)}}{n_\alpha}\psi' + \frac{1}{2}W'^2 + O(\epsilon^4), \tag{4a}$$

$$e_{22}^{(\alpha)} = \gamma_\alpha \frac{\phi}{n_\alpha} + \frac{1}{2}\left(\frac{\psi}{n_\alpha}\right)^2 + O(\epsilon^4), \tag{4b}$$

$$e_{12}^{(\alpha)} = \frac{1}{2}\left(\gamma_\alpha \frac{\psi}{n_\alpha} + W' + \frac{y^{(\alpha)}}{n_\alpha}\phi'\right) + \frac{1}{2}\left[\left(U' + \frac{y^{(\alpha)}}{n_\alpha}\psi'\right)\psi + W'\phi\right]\frac{\gamma_\alpha}{n_\alpha} + O(\epsilon^4), \tag{4c}$$

where the definition $\gamma_\alpha \equiv (-1)^{\alpha+1}$ has been utilized. In order to use (4) for calculation of the average strain energy density, it is convenient to introduce the nondimensional quantities

$$(\bar{U}, \bar{W}, x, \bar{l}) = (U, W, x_1, l)/L, \tag{5a}$$

$$\bar{C}_{ij}^{(\alpha)} = C_{ij}^{(\alpha)}/E_{(m)}, \tag{5b}$$

where the composite is assumed to occupy the domain $-l \le x_1 \le l$, L is a macrodimension to be identified with the wavelength of the buckling pattern and $E_{(m)}$ is a mixture modulus to be defined subsequently. Equations (4, 5) can be used in (1) to obtain the strain energy density of the composite in terms of $(\bar{U}, \bar{W}, \phi, \psi)$ which shall be denoted by q in the sequel. We shall also use Koiter's notation to write the strain energy density of the composite as a sum of homogeneous functionals in q in the form

$$\bar{V}(q) \equiv \frac{1}{E_{(m)}L} \int_{-l}^{l} V \, dx_1$$
$$\equiv V_2(q) + V_3(q) + V_4(q), \tag{6a}$$

wherein the various functionals, on carrying out the x_2-integrations in (1), turn out to be

$$V_2(q) = \frac{1}{2} \int_{-l}^{l} \left\{ E_1 \left(U'^2 + \frac{\epsilon^2}{3} \psi'^2 \right) + G_1 \left(W'^2 + \frac{\epsilon^2}{3} \phi'^2 \right) + E_2\phi^2 + G_2\psi^2 + 2D_1\phi U' + 2D_2\psi W' \right\} dx, \tag{6b}$$

$$V_3(q) = \frac{1}{2} \int_{-l}^{l} \left\{ E_1 U' W'^2 + D_3\phi W'^2 + D_4\phi\psi^2 + G_3\psi^2 U' + 2D_2 W' U' \psi + 2G_2 W' \psi\phi \right\} dx, \tag{6c}$$

$$V_4(q) = \frac{1}{2} \int_{-l}^{l} \frac{1}{4} \{ E_1 W'^4 + D_5\psi^4 + 2D_6\psi^2 W'^2 \} \, dx. \tag{6d}$$

In eqn (6b–d) we have dropped the overbars from the nondimensional quantities defined in (5) for the sake of notational convenience and have used the definitions of composite moduli E_1, G_1, $E_2 \ldots$, etc. given in the appendix.

Equations (6) together with contributions arising from the tractions at the boundaries can be used to write the potential energy functional for the composite, from which the appropriate differential equations for the basic variables denoted by q and the boundary conditions can be derived in a straightforward manner. In this sense the foregoing development is complete as far as the construction of an approximate mathematical model for composite deformation is concerned. The limitation of such a theory, of course, is that only those type of boundary tractions are permissible which are macroscopically homogeneous in the direction normal to the material interfaces. Thus, although it is possible to use the derived model to analyze the composite even for the cases wherein the tractions or the displacements at the boundaries are nonuniform but periodic with the period length being the unit cell dimension, the practically significant situations in which the model is useful are those corresponding to uniform applied boundary conditions. In particular, the theory can be utilized to analyze a laminated medium subjected to uniform compression along the direction of the laminates, and this we shall now do.

MICROBUCKLING ANALYSIS

If the composite is subjected to uniform compressive stress at the boundaries, its potential energy is given by

$$\Pi(q) = V_2(q) + V_3(q) + V_4(q) + \lambda [U]_{-l}^{l}, \tag{7}$$

where λ is the magnitude of the applied compressive traction along the laminates applied at the boundaries, nondimensionalized by the mixture modulus, and

$$[U]_{-l}^{l} \equiv U(l) - U(-l). \tag{8}$$

The potential energy expression (7) shall now be used to obtain: (i) the fundamental equilibrium path, (ii) bifurcation point on the fundamental path and (iii) an asymptotic representation of the bifurcating equilibrium path.

Let the dependent variables along the fundamental path be denoted by $q_0(\lambda)$. If the fundamental path is assumed to be linear, q_0 satisfies

$$V_{11}(q_0, \delta q) + \lambda [\delta U]_{-l}^{l} = 0 \tag{9}$$

where the first term represents the first variation of the functional V_2. The variational statement

(9) reduces to the boundary value problem

$$-E_1U_0''-D_1\phi_0'=0, \tag{10a}$$

$$-\frac{\epsilon^2}{3}G_1\phi_0''+D_1U_0'+E_2\phi_0=0, \tag{10b}$$

$$E_1U_0'+D_1\phi_0=-\lambda;\ \ \frac{\epsilon^2}{3}G_1\phi_0'=0 \quad \text{at } x=\pm l, \tag{10c}$$

where the definition (6b) has been utilized. The corresponding differential equations for W_0 and ψ_0 are homogeneous and have only the trivial solution. Equations (10) yield the fundamental path

$$U_0=-\lambda x/(E_1-D_1^2/E_2), \tag{11a}$$

$$\phi_0=\lambda D_1/E_2\equiv\nu\lambda. \tag{11b}$$

From (11a) it is apparent that $(E_1-E_1^2/E_2)$ is the equivalent modulus for the composite for compression along the laminates. Hence the modulus for scaling in (5b) can be chosen so as to render the nondimensional mixture modulus unity. With this choice, the fundamental path becomes, simply,

$$U_0=-\lambda x,\quad \phi_0=\nu\lambda, \tag{12}$$

and λ, the load parameter, becomes identical to the compressive strain in the composite.

It should be noted here that although the fundamental path (12) has been calculated by neglecting the cubic and quartic functionals in the potential energy expression, it is readily verified that the first variations $V_{21}(q_0,\delta q)$ and $V_{31}(q_0,\delta q)$ vanish, and, therefore, the calculated equilibrium path is an exact solution of the governing equations.

We proceed to conduct the buckling and initial postbuckling analysis by writing the solution of the equilibrium equations in the form

$$q=q_0+Q \tag{13}$$

and calculate the transition potential energy defined by

$$P(Q)=\Pi(q_0+Q)-\Pi(q_0). \tag{14}$$

Let λ_c be a bifurcation point on the fundamental path, to be calculated subsequently. Then, eqns (7) and (12)–(14) yield

$$P(Q)=P_2(Q)+P_3(Q)+P_4(Q)+(\lambda-\lambda_c)P_2'(Q) \tag{15}$$

where

$$P_2(Q)=V_2(Q)+\lambda_cP_2'(Q), \tag{16a}$$

$$P_j(Q)=V_j(Q);\ j=3,4, \tag{16b}$$

and

$$\begin{aligned}P_2'(Q)&\equiv V_{21}(Q,q_0/\lambda)\\ &=-\frac{1}{2}\int_l^l[E_1-\nu D_3)W'^2+(G_3-\nu D_4)\psi^2\\ &\quad+2(D_2-\nu G_2)\psi W']\,dx.\end{aligned} \tag{17}$$

Following Koiter[2], the solution of the variational problem corresponding to (15) will be obtained in the form

$$Q = aq_1 + a^2q_2 + 0[(\lambda - \lambda_c)a, a^3], \tag{18}$$

where a is the amplitude of the buckling mode and q_1, q_2 are solutions of the variational problems

$$P_{11}(q_1, \delta q) = 0, \tag{19}$$

$$P_{11}(q_2, \delta q) + P_{21}(q_1, \delta q) = 0. \tag{20}$$

Equation (19) is the homogeneous problem for the calculation of the buckling mode and λ_c, the value of the load parameter at bifurcation. In writing (20) we have used the result of a subsequent calculation which shows that the quantity $P_3(q_1)$ vanishes so that bifurcation is of the symmetric type in the sense of Koiter[2]. We complete the summary of Koiter's general results by noting that with (18), the transition potential energy of the composite can be written solely in terms of the amplitude a according to

$$F(a) = a(\lambda - \lambda_c)A_2' + a^4A_4 + O(a^5). \tag{21}$$

In (21), which is valid for the special case of symmetric bifurcation, we have used

$$A_2' = P_2'(q_1), \tag{22a}$$

$$A_4 = P_4(q_1) + \frac{1}{2}P_{21}(q_1,q_2). \tag{22b}$$

We shall use the above results for postbuckling analysis for the case in which the composite dimension l is so large that the end effects are negligible. Therefore, it can be assumed that buckling occurs in a sinusoidal pattern whose wavelength can be used for scaling in (5a). As a result, the integrations in the definitions (6b–d) have to be performed over one buckle wavelength which is unity in the scaled system. With these modifications the boundary value problem corresponding to (19) for calculation of bifurcation point and the buckling mode turns out to be

$$-[G_1 - \lambda_c(E_1 - \nu D_3)]W_1'' - [D_2 - \lambda_c(D_2 - \nu G_2)]\psi_1' = 0, \tag{23a}$$

$$-\frac{\epsilon^2}{3}E_1\psi_1'' + [G_2 - \lambda_c(G_3 - \nu D_4)]\psi_1 + [D_2 - \lambda_c(D_2 - \nu G_2)]W_1' = 0, \tag{23b}$$

with periodic boundary conditions at $x = 0, 1$. The corresponding equations for U_1, ϕ_1 have only the trivial solution.

If the solution of (23) is assumed as

$$W_1 = A\cos kx, \tag{24a}$$

$$\psi = kB\sin kx, \quad k = 2\pi, \tag{24b}$$

we obtain from (23) the characteristic equation

$$f\lambda_c^2 + 2\lambda_c g + h = 0, \tag{25}$$

where

$$f = (G_3 - \nu D_4)(E_1 - \nu D_3) - (D_2 - \nu G_2)^2, \tag{26a}$$

$$g = (D_2 - \nu G_2)D_2 - \frac{1}{2}(E_1 - \nu D_3)\left(\frac{\epsilon^2k^2}{3}E_1 + G_2\right)$$

$$-\frac{1}{2}(G_3-\nu D_4)G_1, \tag{26b}$$

$$h=G_1\left(\frac{\epsilon^2k^2}{3}E_1+G_2\right)-D_2^2. \tag{26c}$$

With λ_c as a solution of the characteristic equation, (23)–(24) also yield

$$B=A[G_1-\lambda_c(E_1-\nu D_3)]/[D_2-\lambda_c(D_2-\nu G_2)]. \tag{27}$$

In the sequel we shall use the normalization $A=1$.

To solve the second order problem (20) we first calculate the forcing term in this equation which, on using (24), is given by

$$P_{21}(q_1,\delta q)=k^2\int_0^1\begin{bmatrix}Mk\sin 2kx\delta U\\ +N(1-\cos 2kx)\delta\phi\end{bmatrix}\mathrm{d}x, \tag{28}$$

where

$$M=-\frac{1}{2}(E_1-2D_2B+G_3B^2), \tag{29a}$$

$$N=\frac{1}{4}(D_3-2G_2B+D_4B^2). \tag{29b}$$

We also note here that with $U_1=\phi_1=0$, the coefficient $P_3(q_1)$ vanishes so that bifurcation is of symmetric type as mentioned before. From (20), (28) we obtain the result that W_2 and ψ_2 are trivial and the other components of q_2 satisfy

$$-E_1U_2''-D_1\phi_2'+Mk^3\sin 2kx=0, \tag{30a}$$

$$-\frac{\epsilon^2}{3}G_1\phi_2''+E_2\phi_2+D_1U_2'+N(1-\cos 2kx)k^2=0. \tag{30b}$$

With periodic boundary conditions, the solution of (30) is

$$U_2=kR\sin 2kx, \tag{31a}$$

$$\phi_2=k^2[S\cos 2kx-N/E_2], \tag{31b}$$

where

$$S=\frac{1}{2}(2E_1N+D_1M)\Big/\left[\left(E_2+\frac{4}{3}\epsilon^2k^2G_1\right)E_1-D_1^2\right], \tag{32a}$$

and

$$R=\frac{1}{2}\left[N-\left(E_2+\frac{4}{3}\epsilon^2k^2G_1\right)S\right]\Big/D_1. \tag{32b}$$

We can use the results derived above to calculate the coefficients in the single degree of freedom potential energy functional defined by (21), (22). Thus with (6) and (16), we have

$$A_4=k^4\left[\frac{1}{64}(3E_1+2D_6B^2+3D_5B^4)+\frac{1}{2}\left(\frac{MR}{2}-\frac{N^2}{E_2}-\frac{NS}{2}\right)\right]\equiv\bar{A}_4k^4, \tag{33a}$$

$$A_2' = -\frac{1}{4}k^2[(E_1 - \nu D_3) - 2(D_2 - \nu G_2)B + (G_3 - \nu D_4)B^2]$$
$$\equiv \bar{A}_2 k^2. \tag{33b}$$

Although extremization of the function in (21) can be utilized to obtain the bifurcating path and to answer questions about the stability of this path and about imperfection sensitivity, results of the initial postbuckling analysis for symmetric bifurcation—as is the case here—are somewhat more transparent if obtained in terms of reduction in stiffness of the structure. For this purpose an analysis similar to Budiansky's [12] yields

$$\frac{K_0}{K} = 1 + \frac{1}{2}A_2'^2 K_0 / A_4, \tag{34}$$

which is the ratio of the composite stiffness before buckling to the stiffness immediately after it. With the particular choice of scaling used in this problem, K_0 is unity and, therefore, on using (33), we obtain

$$\frac{K_0}{K} = 1 + \frac{1}{2}\bar{A}_2'^2 / \bar{A}_4. \tag{35}$$

Our analysis of microbuckling of a periodically laminated medium within the context of the approximate continuum model derived here is now complete. We have used the results to conduct a parametric study for the following fixed parameters:

$$\nu_1 = 0.3,\ \nu_2 = 0.45,\ n_1 = 0.6,\ n_2 = 0.4$$

where ν_α denotes the Poisson's ratio for the α constituent. The results of the calculations are shown in Figs. 2–4.

In Fig. 2 the variation of critical strain at buckling with the ratio of the Young's moduli of the constituents has been depicted for the limiting case of infinite buckle wavelength ($\epsilon = 0$). It is evident from the figure that the small strain approximation is valid only for relatively large values of the modulus ratio. A consequence of this result is that for the technically significant metal-matrix composites, microbuckling is likely to occur in the plastic range so that the theory used here is no longer valid for such cases. The same results have been shown in Fig. 3, but

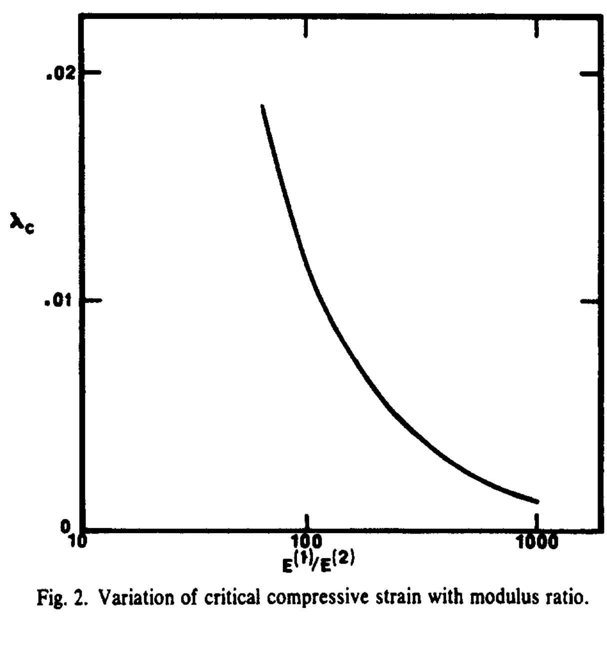


Fig. 2. Variation of critical compressive strain with modulus ratio.

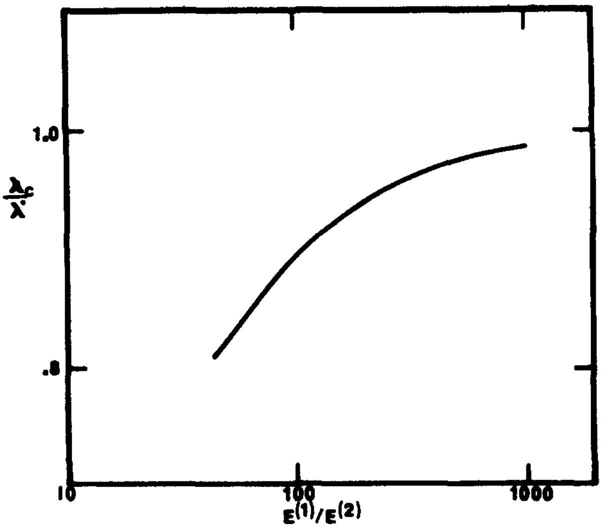


Fig. 3. Variation of critical stress (scaled by composite shear modulus $\lambda^* = 1/(n_1/\mu_1 + n_2/\mu_2)$).

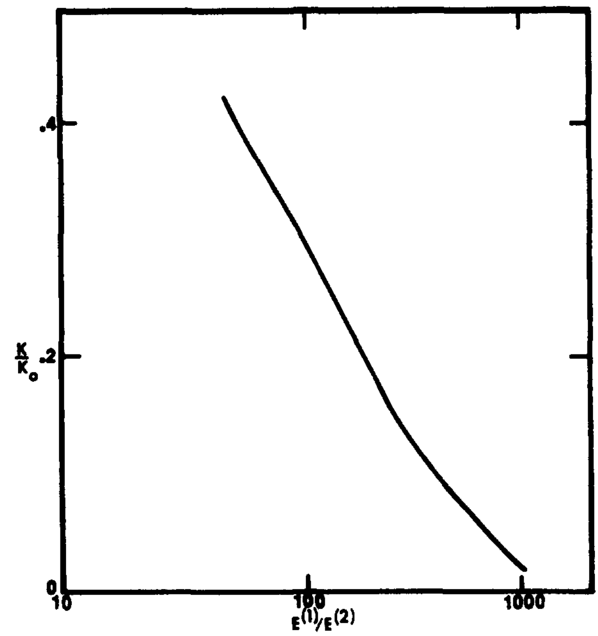


Fig. 4. Variation of the ratio of postbuckling to prebuckling stiffness with modulus ratio.

with buckling stress normalized by $((n_1/\mu_1)+(n_2/\mu_2))^{-1}$ where μ_α denotes the shear modulus. Biot's analysis [3] predicts this parameter to be unity for incompressible media, but the result is seen to be a reasonable approximation for larger values of modulus ratio even without the assumption of incompressibility.

From Fig. 4 it appears that substantial reduction in the stiffness of the composite along the direction of laminates can occur due to microbuckling. However, since the ratio K/K_0 of initial postbuckling to prebuckling stiffnesses is in the range [0, 1], it follows from Budiansky's analysis [12] that the bifurcation is of the stable symmetric type and, therefore, the composite is not imperfection sensitive in that small initial geometrical imperfections are not expected to lead to large decrease in microbuckling stress.

In order to exhibit the effect of finite buckle wavelength, the buckling and initial postbuckling results have been shown for a fixed modulus ratio in Table 1 wherein ϵk is the nondimensional wavenumber of the budkling mode. The trend for microbuckling stress is similar to that of Biot's results and it may be concluded that a more realistic analysis which

Table 1. Effect of buckle wavelength on buckling and initial postbuckling behavior (ratio of Young's moduli $E^{(1)}/E^{(2)} = 100$)

ϵk (Wavenumber)	λ_c (Critical Strain)	K/K_0 (Ratio of Postbuckling-to Prebuckling Stiffness)
0.00	0.0109	0.3253
0.05	0.0112	0.3253
0.10	0.0122	0.3253
0.20	0.0159	0.3252
0.30	0.0220	0.3250

takes into account the end effects in a composite of finite length in the direction of the laminates would predict higher critical stress than the prediction based on infinite wavelength. A somewhat interesting result is that the values of stiffness ratio given in Table 1 do not vary significantly with the wave number of the buckling pattern.

CONCLUDING REMARKS

The results derived on the basis of an approximate continuum model of a periodically laminated medium consisting of linear elastic constituents tend to indicate that compressive microbuckling of the composite can occur at small strain only for those cases wherein the ratio of the moduli of the constituents is relatively large. The analysis of initial postbuckling behavior suggests that the composite is not imperfection sensitive so that the initial waviness of the reinforcements is not expected to reduce the microbuckling stress significantly.

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APPENDIX

Relationships between composite constants and constituent material properties

The composite constants used in (6) and the later development are given in terms of $C_{ij}^{(\alpha)}$—the constituent properties—by the following:

$$E_1 = C_{11}^{(1)} n_1 + C_{11}^{(2)} n_2, \tag{A1}$$

$$E_2 = C_{11}^{(1)}/n_1 + C_{11}^{(2)}/n_2, \tag{A2}$$

$$G_1 = C_{66}^{(1)} n_1 + C_{66}^{(2)}/n_2, \tag{A3}$$

$$G_2 = C_{66}^{(1)}/n_1 + C_{66}^{(2)}/n_2, \tag{A4}$$

$$D_1 = C_{12}^{(1)} - C_{12}^{(2)},\ D_2 = C_{66}^{(1)} - C_{66}^{(2)}, \tag{A5}$$

$$D_3 = D_1 + 2D_2,\ D_4 = \left(\frac{C_{11}^{(1)}}{n_1{}^2} - \frac{C_{11}^{(2)}}{n_2{}^2}\right), \tag{A6}$$

$$D_5 = \frac{C_{11}^{(1)}}{h_1{}^3} + \frac{C_{11}^{(2)}}{n_2{}^3},\quad D6 = \frac{C_{12}^{(1)}}{n_1} + \frac{C_{12}^{(2)}}{n_2}, \tag{A7}$$

$$G_3 = D_6 + 2D_2. \tag{A8}$$