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# THE EFFECT OF MOISTURE ON CARBON FIBER REINFORCED COMPOSITES. IV. PREDICTION OF CHANGES IN THE ELASTIC BEHAVIOR

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UNCLASSIFIED SECURITY CLASSIFICATION OF THIS PAGE (When Date Entered) one can predict the elastic behavior for any \*reasonable\* environment as a function of time. . .. UNCLASSIFIED SECURITY CLASSIFICATION OF THIS PAGE(When Data Entered)

#### SUMMARY

The high cost of modern weapons systems forces increasingly stringent requirements on structural integrity and durability of new military hardware. Prediction of deterioration of material properties in storage and service environments requires therefore an increase in sophistication which necessitates a strong interaction between experiment and analysis.

This report is Part IV of an investigation into the moisture effects in carbon fiber reinforced epoxy composites. Specifically, it deals with the prediction of changes in the elastic behavior of these composites as a result of moisture uptake in a natural environment.

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#### CONTENTS

	Page
INTRODUCTION	1
DISCUSSION	1
CONCLUSIONS	25
ACKNOWLEDGEMENT	25
APPENDIX A	A-1
APPENDIX B	B-1
APPENDIX C	C-1

#### ILLUSTRATIONS

Figure	Title	Page
1	Symmetric Moisture Distributions in a Four Ply Composite	26
2	Effect Of Various Relative Humidities On Resin Modulus (Determined By TBA After 10 Days Exposure. Resin: NARMCO 5208	27
3	Change Of Composite Modulus Ell) as a Function Of Resin Modulus in T 300 CF Unidirectional Composites	28
4	Change Of Composite Modulus (E22) as a Function Of Resin Modulus in T 300 CF Unidirectional Composites	29
5	Change Of Composite Shear Modulus (G12) as a Function Of Resin Shear Modulus in T300 CF Unidirectional Composites.	30
6	Change Of Composite Shear Modulus (G23) as a Function Of Resin Shear Modulus in T300 CF Unidirectional	31
7	Composites Change Of Composite Poisson's Ratio ( $V_{12}$ ) as a Function Of Resin Poisson's Ratio in T 300 CF	
	Unidirectional Composites	32

#### ILLUSTRATIONS (CONT)

#### Figure

Table

and the second se

#### Title

Page

8	Change In Shear Stiffness, Strain At Constant Load	
	(500 lb), and Moisture Uptake of an 8 Ply	
	Laminate 5208/T300 as a Function of Time	
	(Washington DC)	33
9	Arbitrary Moisture Distribution in 8 Ply	
	Laminates	34

#### TABLES

Title

Page

1	Elastic Moduli of Narmco 5208/T300 graphite Fiber lamina and its constituents	17
2	Calculated change of the lamina Young's modulus, E11, of a T300 composite as the matrix modulus $E_m$ changes	18
3	Calculated change of the lamina transverse Young's modulus, E22, of a T300 composite as the matrix modulus changes	18
4	Calculated change of the lamina longitudinal shear modulus, G12, of a T300 composite as the matrix shear modulus, Gm, changes	19
5	Calculated change of the lamina transverse shear modulus, G <sub>23</sub> , of a T300 composite as the matrix shear modulus G <sub>m</sub> changes	19
6	Calculated change in the lamina major Poisson's ratio, $v_{12}$ , of a T300 composite as the matrix Poisson's ratio $v_m$ changes	20
7	Young's and Shear Moduli of Narmco 5208 Resin Equilibrated at Various Relative Humidities	20
8	"A Matrices" for 8 ply T300/Narmco 5208 (Vf = .7) laminates ( $\theta$ = 0° and $\theta$ = +45° symm.) after various exposure times to Washington, D.C.	20
	weather conditions	22
9	Effect of Internal Moisture Distribution on the Elastic Behavior of Composites	24

#### INTRODUCTION

This report is part IV of an investigation into the effects that moisture has on carbon fiber-reinforced epoxy composites. Part I discussed the diffusion of moisture in composites, part II summarized our experimental observations on the reversible changes in strengths of composites after moisture exposure, part III described an analytical modeling of uptake and internal distribution of moisture in composites with respect to real outdoor weathering conditions. This report discusses the changes in the elastic behavior of composite as a result of outdoor exposure.

A unified scheme for the prediction of the elastic behavior of multi-angleplied laminates is presented. It includes temperature effects, swelling effects and changes in the state of stress (strain) of a laminated composite as a function of time under any prescribed climatic environment.

#### DISCUSSION

#### A. Problem

It has been observed by various investigators [1-7] that exposure of fiber-reinforced epoxy composites to moisture leads to a reduction in matrix dominated strength properties. The degree of

- [1] J. Hertz, Final Report NASA 8-27435 Jmrs 1973 (NASA Contract)
  [2] C. E. Browning, 28th Annual Conference of the Society of
- Plastic Industry, Feb 1973 (Washington, D.C.) Proceedings 15A.
- [3] J. M. Augl and A. E. Berger, "Moisture Effects on Carbon Fiber Epoxy Composites". <u>Proceedings p. SAMPE Meeting</u>, Seattle, Washington, Oct 1976.
- Washington, Oct 1976.
   J. M. Augl, "The Effect of Moisture on Carbon Fiber-Reinforced Epoxy Composites. II Mechanical Property Changes." NSWC/WOL/TR 76-149 (1976).
- [5] D. H. Kaelble, P. J. Dynes and L. Maus, "Hydrothermal Aging of Composite Materials Part 1: Interfacial Aspects." Journal of Adhesion 8121 (1976) Part 2: Matrix Aspects ibid 8, 155 (1976).
- [6] E. L. McKague, J. E. Halkias and J. D. Reynolds "Moisture in Composites. The Effect of Supersonic Service on Diffusion." J. Comp. Mat. 9, 2 (1975).
- [7] Proceedings Air Force Workshop on Durability Characteristics of Resin Matrix Composities. Battelle Columbus Laboratories, Sept 30, 1975.

strength reduction depends, of course, on the type of failure mechanism, on the moisture concentration, and on the temperature. The strength degradation is reversible, i.e., after removal of the absorbed moisture the original strength can be restored. Also, the strength loss levels out after the equilibrium concentration with the environment has been reached [4].

In Part I of this report [8] we have discussed the diffusion of moisture in composites and in Part III [9] we have described an analysis of how real outdoor and service conditions affect the uptake and the internal moisture distribution in composites.

In the following we shall discuss how a real outdoor and service environment affects the elastic properties of carbon fiber reinforced epoxy composites, i.e., the change of the elastic properties as a function of time in a prescribed climatic environment.

We shall restrict ourself to laminated plates that are composed of transversely isotropic unidirectional plies layed up at different fiber-angles. We also assume that the plate is exposed to moisture on both surfaces.

An example of such a composite is shown in Figure 1 which gives a 4 ply, +450, symmetric laminate. The curved line (shaped like a parabola) indicates a superposed profile of moisture concentration after a certain time of moisture exposure which is given by the solution of the diffusion equation as discussed in reference [8] and **D**]. The moisture distribution is symmetric, however, the individual plies are no longer transversely isotropic nor are they symmetric because the elastic moduli of the matrix is not constant anymore in the z-direction since the modulus of the matrix is dependent on the moisture concentration [4]. We can, therefore, no longer take the individual elastic constants for the laminae that constitute the laminate but we have to know the change of the lamina elastic constants as a function of absorbed moisture, and, if we are interested in the temperature effects, we have also to determine the lamina elastic constants as a function of temperature.

However, we can approximate this continuous change in the matrix modulus in the z-direction by finite, discontinuous, steps ( $\Delta$ ) "sufficiently small" to neglect the concentration differences on either side of the  $\Delta$ -layers, and we consider these  $\Delta$ -layers again as having transversely isotropic symmetry. What is "sufficiently small" is a question of the change in modulus with moisture concentration and the sensitivity of such measurements. The  $\Delta$  differences should depend on the gradient of the moisture concentration, and are equal or smaller than the physical plies.

[8] J. M. Augl and A. E. Berger, "The Effect of Moisture on Carbon Fiber-Reinforced Composites. I Diffusion." NSWC/WOL/TR 76-7 (1976).

[9] J. M. Augl and A. E. Berger, "The Effect of Moisture on Carbon Fiber-Reinforced Composites; III Prediction of Moisture Sorption in a Real Outdoor Environment." NSWC/WOL/TR 77-13 (1977).

Thus, the whole problem reduces again to classical laminated plate theory, which has been amply described in various papers and monographs of which we want to cite only a few [10 - 14].

This theory may be expressed by the compact matrix equation (1)

 $\begin{bmatrix} \underline{N} \\ \overline{M} \end{bmatrix} = \begin{bmatrix} \underline{A} \\ \overline{B} \\ \overline{D} \end{bmatrix} \begin{bmatrix} \underline{\varepsilon}^{O} \\ \overline{\kappa} \end{bmatrix}$ (1)

where the N matrix is the stress resultant matrix; M is the moment resultant matrix;  $\varepsilon^{O}$  is the matrix describing the midplane stretching and shearing deformations, and  $\kappa$  describes twisting and bending deflections; the A, B, and D matrices are the extensional stiffness matrix, the bending-stretching coupling matrix and the flexural stiffness matrix respectively.

The following steps are therefore required:

1. Determine the moisture diffusion coefficient of the composite in the z-direction as a function of temperature (and, if necessary, as a function of concentration).

2. Calculate the internal moisture concentration distribution for a prescribed environment (with variable temperature and humidity) as a function of time by solving Fick's second diffusion equation.

3. Determine the constitutive properties  $Q_{ij}$  of each  $\Delta$  - layer as a function of uniform moisture distribution and as a function of temperature.

4. Calculate the stiffness matrix of the laminate (A, B, D) from the corresponding  $\Delta$  -  $Q_{i\,j}$  .

We have discussed step 1 and 2 previously [8, 9]. In the following we shall investigate step 3 and step 4.

[10]	J. E. Ashton, J. C. Halpin and P. H. Petit, "Primer on Composite
	Materials." Composite Materials Workshop, Technomic Publ.
- 7	Co., Inc., Stanford, Conn. (1969).
[11]	J. C. Halpin and S. W. Tsai, "Environmental Factors in Composite
- 7	Materials Design" AFML TR 67-423.
[12]	J. R. Vinson and T. W. Chou, "Composite Materials and their
	Use in Structures" John Wiley 1975.
[13]	S. W. Tsai, "Mechanics of Composite Materials. Part II."
	AFML-TR 149 (1966).
[14]	S. G. Lekhnitskii, "Anisotropic Plates." Translated by
	S. W. Tsai and T. Cheron, Gordon and Breach, 1968.

#### B. <u>A-Layer Constitutive Properties</u>

For an orthotropic lamina oriented in the principal material direction, such that the fibers are aligned parallel to the l-axis as shown by the sketch below



the three dimensional constitutive relations are

[01 ]		Q <sub>11</sub>	Q <sub>12</sub>	Q <sub>13</sub>	0	0	0	ΙΓεη
σ2		Q <sub>11</sub> Q <sub>12</sub>	Q22	Q <sub>23</sub>	0	0	0	ε
σ3	=	Q <sub>13</sub>	Q <sub>22</sub> Q <sub>23</sub>	Q <sub>33</sub>	0	0	0	ε <sub>1</sub> ε <sub>2</sub> ε <sub>3</sub>
τ23		0 0	0	0	2Q <sub>44</sub>	0	0	12423
τ <sub>13</sub>		0	0	0	0	20 <sub>55</sub>	0	12Y13
$ \begin{matrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \\ \tau_{23} \\ \tau_{13} \\ \tau_{12} \end{matrix} $		0	0	0	0	0	0 20 <sub>66</sub>	<sup>2</sup> Υ <sub>23</sub> <sup>2</sup> Υ <sub>13</sub> <sup>2</sup> Υ <sub>12</sub>

where  $\sigma_1$ ,  $\sigma_2$ ,  $\sigma_3$ ,  $\tau_{23}$ ,  $\tau_{13}$ ,  $\tau_{12}$  are the normal and shear stresses,  $\varepsilon_1$ ,  $\varepsilon_2$ ,  $\varepsilon_3$ ,  $\gamma_{23}$ ,  $\gamma_{13}$ ,  $\gamma_{12}$  are the corresponding strains and the  $Q_{ij}$ are the components of the stiffness matrix.

We have used the momenclature of reference [10, 12]. Since we can assume that the fibers are parallel to the 1 axis but randomly distributed in the plane perpendicular to the 1 axis, this 2-3-plane is a plane of isotropy and the lamina is said to be "transversely isotropic" or sometimes also called "monotropic." For such a system the following relations hold:

$$Q_{33} = Q_{22}, Q_{13} = Q_{12}, Q_{55} = Q_{66}$$
  
and  $Q_{44} = \frac{1}{2} (Q_{22} - Q_{23})$ 

Further, in thin plate and shell theory, we assume that there are no stresses applied in the z-direction (= 3-direction) therefore  $Q_{13} = Q_{23} = Q_{33} = 0$ .

A detailed discussion of the elastic stiffness matrices for all the crystallographic symmetry classes was given by S. G. Leckhnitskii [15] (Attention should be given to the confusion that may arise from naming the axes 1, 2, 3, and z, y, z by different authors with respect to fiber direction and plane of isotropy.)

The physical meaning of the Q<sub>ij</sub> matrix elements are as follows:

$$Q_{11} = \frac{E_{11}}{(1 - v_{12} v_{21})}$$
(3)

$$Q_{12} = \frac{E_{22}}{(1 - v_{12} v_{21})}$$
(4)

$$Q_{12} = Q_{21} = \frac{v_{21} E_{11}}{(1 - v_{12} v_{21})} = \frac{v_{12} E_{22}}{(1 - v_{12} v_{21})}$$
(5)

 $Q_{44} = G_{23}$ 

 $Q_{55} = Q_{66} = G_{12}$ 

Where  $E_{11}$  and  $E_{22}$  are the Young's moduli in the 1 and 2 directions and  $G_{12}, G_{23}$  are the longitudinal and transverse composite shear moduli, and  $v_{12}$  is the principal Poisson's ratio.

[15] S. C. Lekhnitskii, <u>Theory of Elasticity of Anisotropic</u> Elastic Bodies. Holden Day 1963.

If the stiffnesses of a plate are to be determined for some new reference axes (x, y) in the plane of the lamina, this can be accomplished by a matrix transformation describing a rotation of the coordinates around the 3-axis (= z-axis) by an angle\*  $\theta$  as indicated by 6-8.

$$[\sigma]_{(xy)} = [T]^{-1} [\sigma]_{(12)}$$
(6)
$$[\varepsilon]_{(xy)} = [T]^{-1} [\varepsilon]_{(12)}$$
(7)

(8)

and thus

$$[\sigma]_{(xy)} = [T]^{-1} [Q_{ij}] [T] [\varepsilon]_{(xy)}$$

where

$$[\mathbf{T}] = \begin{bmatrix} m^2 n^2 0 & 0 & 2mn \\ n^2 m^2 & 0 & 0 & -2mn \\ 0 & 0 & m-n & 0 \\ 0 & 0 & n & m & 0 \\ -mn & mn & 0 & 0 & (m^2 - n^2) \end{bmatrix}$$

and

$$\begin{bmatrix} \mathbf{T} \end{bmatrix}^{-1} = \begin{bmatrix} m^2 & n^2 & 0 & 0 & -2mn \\ n^2 & m^2 & 0 & 0 & 2mn \\ 0 & 0 & m & n & 0 \\ 0 & 0 & -n & m & 0 \\ mn & -mn & 0 & 0 & (m^2 - n^2) \end{bmatrix}$$

and where  $m \equiv \cos \theta$ ,  $n = \sin \theta$ .

\*The angle is (+) if it is measured counterclockwise from the axis to the (+1) axis.

Thus a stiffness matrix  $(\overline{Q}_{ij})$  is obtained describing a generally orthotropic lamina. Stress strain relations for such a generally orthotropic lamina including transverse shear deformation are given by (9)



where  $\left[\overline{Q}_{ij}\right] = \left[T\right]^{-1} \left[Q_{ij}\right] \left[T\right]$ 

The evaluation of the  $\overline{Q}_{ij}$  was simplified by Tsai and Pagano [16] who used certain functions that are independent of  $\theta$  and are given by the following relations

$$\begin{split} \overline{Q}_{11} &= U_1 + U_2 \cos (2\theta) + U_3 \cos (4\theta) \\ \overline{Q}_{22} &= U_1 - U_2 \cos (2\theta) + U_3 \cos (4\theta) \\ \overline{Q}_{12} &= U_4 - U_3 \cos (4\theta) \\ \overline{Q}_{66} &= U_5 - U_3 \cos (4\theta) \\ \overline{Q}_{16} &= -\frac{1}{2} U_2 \sin (2\theta) - U_3 \sin (4\theta) \\ \overline{Q}_{26} &= -\frac{1}{2} U_2 \sin (2\theta) + U_3 \sin (4\theta) \end{split}$$

where

$$U_{1} = \frac{1}{8} (3 Q_{11} + 3 Q_{22} + 2Q_{12} + 4Q_{66})$$

$$U_{2} = \frac{1}{8} (Q_{11} - Q_{22})$$

$$U_{3} = \frac{1}{8} (Q_{11} + Q_{22} - 2Q_{12} - 4Q_{66})$$

$$U_{4} = \frac{1}{8} (Q_{11} + Q_{22} + 6Q_{12} - 4Q_{66})$$

$$U_{5} = \frac{1}{8} (Q_{11} + Q_{22} - 2Q_{12} + 4Q_{66})$$

[16] S. W. Tsai and N. Pagano, "Invariant Properties of Composite Materials." <u>Composite Materials Workshop</u>. Technomic Publ. Co., Inc., pp. 233-53 (1968).

(10)

(11)

7

and the quantities  $\overline{Q}_{44}$   $\overline{Q}_{55}$   $\overline{Q}_{45}$  were given by J. R. Vinson and T. W. Chou [12],

 $\overline{Q}_{44} = Q_{44} \cos^2 \theta + Q_{55} \sin^2 \theta$   $\overline{Q}_{55} = Q_{44} \sin^2 \theta + Q_{55} \cos^2 \theta$  $\overline{Q}_{45} = (Q_{55} - Q_{44}) \cos \theta \sin \theta$ 

The determination of the required  $\Delta$ -layer constitutive properties can be either measured directly on unidirectional composite specimens or predicted from the constituent properties.

a. Direct measurement of  $\Delta$ -layer constitutive properties.

To obtain all the elastic constants of a lamina for various levels of moisture and temperatures is a rather involved experimental program.

In order to prepare the test specimens it is necessary to expose the samples at various constant relative humidity levels for a long enough time to reach a uniform moisture concentration. For a .25 cm thick composite this may take about 80 days even at 75°C. The following measurements of composite elastic constants will have to be carried out at about 5 different temperatures (between room temperature and 170°C) and 5 different humidity levels: Ell, E22, G12, v12, G23. If only a minimum of 5 specimens are used per condition, this would amount to 625 specimens.

b. Prediction, of  $\Delta$ -Layer Constitutive Properties from the resin and fiber properties.

Numerical techniques have been developed to predict the composite elastic constants from the known properties of the matrix and fiber. These problems can be solved rigorously if a periodic, geometric, array of fibers is assumed (for instance, tetragonal or hexagonal). Also, for a statistical distribution of fibers, solutions have been described.

Even if the prediction of properties is not always very accurate, these formulas are quite useful for our purpose of predicting changes of properties.

The reason why the theories do not always predict accurately the experimentally determined values may be due to the fact that one or several of the assumptions made for the model are not correct. Most of these theories make the following assumptions:

1. The ply is macroscopically homogeneous, linearly elastic, and transversely isotropic.

(12)

2. The fibers are linearly elastic and homogeneous.

3. The matrix is linearly elastic and homogeneous.

Fiber and matrix are free of voids.

5. There is complete bonding at the interface between fiber and matrix and there is no transition region.

6. The fiber and matrix are initially in a stress free state.

7. The fibers are regularly spaced and aligned.

In a real composite neither assumption is totally justified. Composites have voids, the fibers are not uniform, they are not regularly spaced, they are not perfectly aligned, there is not everywhere a perfect bound between resin and fiber, there is experimental (microscopic) evidence that there is a transition zone between resin and fiber, simple thermal calculation show that there may be considerable residual stresses in composites (with high Tg resins) that have been cured at a high temperature. It is therefore rather surprising how well the models predict the composite properties.

What we are interested in here is not so much the absolute values of the elastic constants but rather the changes in properties.

Even, if the assumptions are true only to a certain extent, the violation will remain relatively constant if we let moisture permeate into the resin. There will be no change in the number of voids; the relative fiber spacings remains constant; the misalignment remains unchanged; the transient zone remains, although it may change properties; the interfacial bonding may be weakened, this is particularly true for glass fiber composites; moisture, however, may enhance relaxation of residual stresses.

Thus, after the initial composite unidirectional properties have been experimentally determined in the dry state, as baseline properties, we expect that the relative changes in the elastic composite properties, due to moisture sorption, can be predicted with a considerably higher degree of accuracy than it can be done from original resin and fiber constituent properties, so that it is only necessary to determine the matrix elastic constants  $E_m$ ,  $G_m$  (or  $v_m$ ) as a function of uniform moisture concentration and temperature.

These properties can be determined nondestructively on very few simple plate specimens by ultrasonic sound velocity measurements of longitudinal and shear waves. Torsional pendulum experiments combined with a few tensile measurements may also be used. Since there is a certain moisture desorption at the high temperature measurements, it is preferable to use not too thin plate specimens (.3 cm thickness will probably suffice).

We made no attempt here to review the various approaches that have been taken to predict the elastic constants of unidirectional composites but rather list in Appendix A a number of solutions for the different elastic constants that have proved to be quite useful in predicting these properties.

#### C. Properties of a Laminate with Absorbed Moisture.

After moisture has penetrated the composite, the modulus of the matrix is not only changed in the neighborhood of the glass transition temperature  $(T_q)$  but over a very wide temperature range (see Figure 2).

Since the matrix modulus enters in all the prediction formulas of the composite elastic constants (see Appendix A), it is obvious that the composite properties change accordingly. If we know from experiment the degree of change of the matrix with moisture and temperature and if we know the internal moisture distribution, we can describe in a straight forward fashion the composite elastic behavior, by using the same formalism as is used for analyzing laminates with a homogeneous matrix and different ply-orientations. Instead of treating the laminate as being composed of real plies we describe it in terms of the above mentioned artificial  $\Delta$ -layers, and we simply rewrite the equations of laminated plate theory in terms of  $\Delta$ -layers as if the composite were composed of a hybride of  $\Delta$ -layers. In the following we adapt the basic nomenclature used in reference [10, 12].

$$\left[\sigma\right]_{k\Delta} = \left[\overline{Q}\right]_{k\Delta} \left[\varepsilon\right] \tag{13}$$

where  $\sigma$  and  $\varepsilon$  are the stress and strain matices and  $\overline{Q}$  is the stiffness matrix with respect to the laminate axes (x,y,z), the subscript k $\Delta$  refers to the k<sup>th</sup>  $\Delta$ -layer. This equation can be rewritten in terms of strain-displacement relations

$$\left[ \varepsilon \right]_{k\Delta} = \left[ \varepsilon^{\circ} \right] + z \left[ \kappa \right]$$
 (14)

15

where

$$\varepsilon_{\mathbf{x}}^{\mathbf{o}} = \frac{\partial \mathbf{u}_{\mathbf{o}}}{\partial \mathbf{x}}; \ \varepsilon_{\mathbf{y}}^{\mathbf{o}} = \frac{\partial \mathbf{v}_{\mathbf{o}}}{\partial \mathbf{y}}; \ \varepsilon_{\mathbf{x}\mathbf{y}}^{\mathbf{o}} = \frac{1}{2}(\frac{\partial \mathbf{u}_{\mathbf{o}}}{\partial \mathbf{y}} + \frac{\partial \mathbf{v}_{\mathbf{o}}}{\partial \mathbf{x}})$$

 $\kappa_{\mathbf{x}} = -\frac{\partial^2 w}{\partial \mathbf{x}^2}$ ;  $\kappa_{\mathbf{y}} = -\frac{\partial^2 w}{\partial \mathbf{y}^2}$  and  $\kappa_{\mathbf{x}\mathbf{y}} = -2 \frac{\partial^2 w}{\partial \mathbf{x} \partial \mathbf{y}}$ 

and

substituting (14) into (13) gives

$$\left[\sigma\right]_{k\Delta} = \left[\overline{\varrho}\right]_{k\Delta} \left[\varepsilon^{\circ}\right] + z \left[\overline{\varrho}\right]_{k\Delta} \left[\kappa\right]$$
 (15)

The stress and momentum resultants can be written in the usual way by

$$\begin{bmatrix} N_{x} \\ N_{y} \\ N_{xy} \\ Q_{x} \\ Q_{y} \end{bmatrix} = \int_{-h/2}^{h/2} \begin{bmatrix} \sigma_{x} \\ \sigma_{y} \\ \tau_{xy} \\ \tau_{xz} \\ \tau_{yz} \end{bmatrix} dz, \begin{bmatrix} M_{x} \\ M_{y} \\ M_{xy} \end{bmatrix} = \int_{-h/2}^{h/2} \begin{bmatrix} \sigma_{x} \\ \sigma_{y} \\ \sigma_{xy} \end{bmatrix} z dz$$
(16)

where the N<sub>x</sub> N<sub>y</sub> N<sub>xy</sub> are the in-plane stress resultants, M<sub>x</sub> M<sub>y</sub> M<sub>xy</sub> are the stress couples.  $Q_x$  and  $Q_y$  are the transverse shear forces and h is the thickness of the laminate.

Summation over all  $\Delta$ -layers yields

$$\begin{bmatrix} N_{x} \\ N_{y} \\ N_{xy} \end{bmatrix} = \sum_{k\Delta = 1}^{n} \int_{h_{k\Delta - 1}}^{h_{k\Delta}} \begin{bmatrix} \sigma_{x} \\ \sigma_{y} \\ \tau_{xy} \end{bmatrix} dz$$

$$= \sum_{k\Delta = 1}^{n} \int_{h_{k\Delta - 1}}^{h_{k}\Delta} \left[ \overline{Q} \right]_{k\Delta} \begin{bmatrix} \varepsilon_{x}^{o} \\ \varepsilon_{y}^{o} \\ \varepsilon_{xy}^{o} \end{bmatrix} dz + \int_{h_{k\Delta - 1}}^{h_{k}\Delta} \overline{Q} k\Delta \begin{bmatrix} \kappa_{x} \\ \kappa_{x} \\ \kappa_{x} \\ \frac{1}{2}\kappa_{xy} \end{bmatrix} zdz$$
(17)

since  $[\epsilon^{\circ}]$  and  $[\kappa]$  are not functions of z, one can write (17) as

$$\begin{bmatrix} N_{\mathbf{x}} \\ N_{\mathbf{y}} \\ N_{\mathbf{x}\mathbf{y}} \end{bmatrix} = \sum_{k_{\Delta} = 1}^{n} \left\{ \begin{bmatrix} \overline{Q} \end{bmatrix}_{k_{\Delta}} \begin{bmatrix} \varepsilon^{O} \end{bmatrix} \int_{h_{k\Delta} - 1}^{h_{k\Delta}} dz + \begin{bmatrix} \overline{Q} \end{bmatrix}_{k_{\Delta}} \begin{bmatrix} \kappa \end{bmatrix} \int_{h_{k\Delta} - 1}^{h_{k\Delta}} z dz \right\}$$
(18)

or in short form

$$\begin{bmatrix} N \end{bmatrix} = \begin{bmatrix} A \end{bmatrix} \begin{bmatrix} \varepsilon^{\circ} \end{bmatrix} + \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} \kappa \end{bmatrix}$$
(19)

where

$$A_{ij} = \sum_{k\Delta = 1}^{n} (\bar{Q}_{ij})_{k\Delta} (h_{k\Delta} - h_{k\Delta-1})$$
(20)

$$B_{ij} = \sum_{k\Delta = 1}^{n} (Q_{ij})_{k\Delta} (h^{2}_{k\Delta} - h^{2}_{k\Delta - 1})/2$$
(21)  
(i, j = 1, 2, 6)

Similarly for

$$\begin{bmatrix} M_{\mathbf{x}} \\ M_{\mathbf{y}} \\ M_{\mathbf{x}\mathbf{y}} \end{bmatrix} = \int_{-h/2}^{h/2} \begin{bmatrix} \sigma_{\mathbf{x}} \\ \sigma_{\mathbf{y}} \\ \tau_{\mathbf{x}\mathbf{y}} \end{bmatrix} z dz = \sum_{k\Delta = 1}^{n} \int_{k_{k\Delta - 1}}^{k_{k\Delta}} \begin{bmatrix} \sigma_{\mathbf{x}} \\ \sigma_{\mathbf{y}} \\ \tau_{\mathbf{x}\mathbf{y}} \end{bmatrix} z dz$$

$$= \left\{ \sum_{k_{\Delta}=1}^{n} \left[ \overline{Q} \right]_{k_{\Delta}} \int_{h_{k_{\Delta}-1}}^{h_{k_{\Delta}}} z dz \right\} \left[ \varepsilon^{\circ} \right] + \left\{ \sum_{k_{\Delta}=1}^{n} \left[ \overline{Q} \right]_{k_{\Delta}} \int_{h_{k_{\Delta}-1}}^{h_{k_{\Delta}}} z^{2} dz \right\} \left[ \varepsilon^{\circ} \right]$$

$$[M] = [B] [\varepsilon^{\circ}] + [D] [\kappa]$$
(22)

where

$$D_{ij} = \frac{1}{3} \sum_{k\Delta = 1}^{n} \left[ \overline{Q}_{ij} \right]_{k\Delta} \begin{pmatrix} h_{k\Delta}^{3} - h_{k\Delta - 1}^{3} \\ (i, j = 1, 2, 6) \end{pmatrix}$$
(23)

if the transverse shear forces are to be included the assumption is made that the transverse shear stresses are distributed parabolically across the laminate thickness and a weighing function as used by Reissner [17] was employed

$$f(z) = \frac{5}{4} \left[ 1 - \left( \frac{z}{h/2} \right)^2 \right]$$
(24)

the transverse shear resultants are obtained for the  $k^{th} \Delta$  - layer

$$\tau_{xz}^{(k\Delta)} = 2 \ \overline{Q}_{55}^{(k\Delta)} \ \frac{1}{2} \gamma_{xz} + 2 \ \overline{Q}_{45}^{(k\Delta)} \ \frac{1}{2} \gamma_{yz}$$

$$\tau_{yz}^{(k\Delta)} = 2 \ Q_{45}^{(k\Delta)} \ \frac{1}{2} \gamma_{xz} + 2 \ Q_{44}^{(k\Delta)} \ \frac{1}{2} \gamma_{yz}$$
(25)

and from (16) one obtains

$$Q_{x} = 2 \left( A_{55} \frac{1}{2} \gamma_{xz} + A_{45} \frac{1}{2} \gamma_{yz} \right)$$

$$Q_{y} = 2 \left( A_{45} \frac{1}{2} \gamma_{xz} + A_{44} \frac{1}{2} \gamma_{yz} \right)$$
(26)

where

$$A_{ij} = \frac{5}{4} \sum_{k\Delta=1}^{n} \left( \overline{Q}_{ij} \right)_{k\Delta} \left[ h_{k\Delta-1} - \frac{4}{3} \frac{(h_{k\Delta}^{3} - h_{k\Delta-1}^{3})}{h^{2}} \right]$$
(27)  
(i, j = 4, 5)

Finally the inclusion of all artificial  $\Delta$ -layers leaves the overall formalism unchanged and it can be written as is customary by



(28)

and by (26), or by the abbreviated formula (1) which comprise the full system of constitutive equations to study laminates that are symmetrically permeated by moisture. Other useful relations between N, M, A, B and D are given in Appendix B.

The changes with time are governed by the diffusion equation.

#### D. Property Changes in Laminates due to the Effect of Temperature On the Matrix Modulus

The matrix modulus changes with temperature and, increasingly so, as the Tg of the matrix is approached.

Again, the formalism discussed in the previous sections remains the same.

The Fick's diffusion equation has the same form also for thermal diffusion. The only difference is that the thermal diffusivity is several orders of magnitude larger than the moisture diffusion. The thermal diffusion is so much faster that for most problems in composite structures we are more interested in the isothermal steady state effects at different temperatures, or with constant temperature gradient rather than the transient phenomena of non-steady state heat Though there is no difficulty in dealing with transient flux. conditions because of the difference in rates of thermal and moisture diffusion. The changes in the elastic behavior due to temperature changes and transient internal heat fluxes is simply superposed on the "constant" internal moisture distributions. Even the same  $\Delta$ -layer spacings may be used. The changes of the properties with time are now governed by the thermal diffusion equation, which requires that the thermal diffusivity of the composite in the z-direction be known.

The matrix elements of the stiffness tensor  $(\overline{Q}_{ij})_{k\Delta}$  are replaced by the elements  $(\overline{Q}_{ij} (C_{H20},T))_{k\Delta}$  indicating that the  $\Delta$  layer elastic constants depend on internal moisture concentration and temperature.

## E. Property Changes in a Laminate due to Volume changes caused by solvent swelling or temperature changes.

In addition to affecting the modulus of the matrix, moisture sorption and temperature changes have similar effects in that they change the specific volumina of the constituent materials unequally. This leads to a change in the internal state of stress.

We do not intend to discuss this subject here further since it was well described by Pipes, Vinson and Chou in a recent paper [18].

[18] R. B. Pipes, J. R. Vinson and T. W. Chou, "On the Hygrothermal Response of Laminated Composite Systems." <u>J. Comp.</u> <u>Materials</u> 10, 129 (1976).

These authors have modified the constitutive equations of the laminated plate theory to include these effects. And, for a given laminate strain, the stresses within each lamina were given by

$$\sigma_{j}^{k}(z,t) = Q_{ij}^{k} \left[ \varepsilon_{j}^{0}(t) + z \kappa_{j}(t) - \alpha_{j}^{k} T(z,t) - \beta_{j}^{k} M(z,t) \right]$$
(i, j = 1,2,6)

where  $\sigma_j$ ,  $Q_{ij}$ ,  $\varepsilon_j^o$  and  $\kappa_j$  are the stresses, stiffness components, inplane strains and bending strains respectively;  $\alpha_j$  and  $\beta_j$  are the thermal and hygroscopic expansion coefficients and T(z,t) and M(z,t)are the temperature and moisture concentration distributions, at the time t. The superscript k refers to the k<sup>th</sup> ply.

This formula still neglects the temperature and humidity dependence of the matrix modulus.

#### F. Combined Effects of Temperature and Moisture on the Elastic Behavior of Composites

Expanding on the effort of Pipes, Vinson and Chou we propose to modify their equation (29) to include the changes in the matrix modulus and therefore in the  $\Delta$ -layer stiffness components to the following form:

$$\begin{bmatrix} \sigma_{j}(T, C_{H_{2}0}, t) \end{bmatrix}_{k\Delta} = \begin{bmatrix} \overline{Q}_{ij}(T, C_{H_{2}0}, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) T(z, t) - \beta_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(t) - \alpha_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix} \varepsilon_{j}^{o}(t) + z\kappa_{j}(k\Delta) M(z, t) \end{bmatrix}_{k\Delta} \begin{bmatrix}$$

where the symbols are the same as in equation 29, however the  $(\overline{Q}_{i\,j})$ 's are now functions of temperature, moisture concentration and time, whereby the concentration of moisture, CH2O, and the time t are related by the Fick's diffusion equation, and the temperature and time are similarly related by the analogous equation for heat conduction. However, because of the large rate differences in thermal and moisture diffusion, it is more convenient to treat these two phenomena separately.

#### G. <u>Computer Program for Calculating the Elastic Stiffness Matrix</u> of a Laminate as a Function of a Real Outdoor Environment

For calculating the change in the elastic behavior of a composite for real outdoor environment we require the following input data:

(29)

1. Long-term average temperature and relative humidity of the environment, or 1-3 hourly weather data of past years on magnetic tape.

2. Diffusion coefficient of the laminate in the z-direction as a function of temperature (and concentration, if necessary), or the diffusion coefficient of the matrix as a function of temperature (and concentration) together with the composite fiber volume fraction.

3. Equilibrium concentration of the matrix material as a function of relative humidity (and temperature, if necessary).

4. Fiber and matrix moduli ( $E_m$  and  $G_m$  or  $v_m$ ,  $E_{11f}$ ,  $E_{22f}$ ,  $G_{12f}$ ,  $v_{12f}$ ) as a function of uniform moisture concentration and temperatue. (The fiber properties are considered to remain constant over the temperature range of interest.) For predicting the lamina properties  $E_{11}$   $E_{12}$ ,  $G_{12}$  and  $v_{12}$  the Halpin-Tsai equations (Al7-Al9) were used and for  $G_{23}$  Rosen's Equation (Al6) was used.

5. Laminate geometry (number of plies, orientation of plies, thickness of plies, thickness of  $\Delta$ -layers).

6. Time intervals of printout of the stiffness matrix.

Applied laminate stresses or strains (σj,εj).

The printout for the specified time intervals gives:

1. The  $Q_{ij}^{K\Delta}$  (if desired)

2. The matrices A, B, D, A\*, B\*, C\*, D\*

3. The strains and stresses in each lamina  $(\epsilon_j^k, \sigma_j^k)$  or, if desired of each  $\Delta$ -layer.

H. Examples.

Example 1: Composite stiffness changes as a function of matrix stiffness changes.

Moisture will plasticize the matrix and reduce the values of the matrix elastic properties depending on the amount of (uniformly distributed) moisture and on the temperature.

Let us assume that these values have been determined experimentally, what is the corresponding change in the lamina elastic constants.

Table 1 gives the original fiber matrix and lamina properties. Table 2-6 and Figures 3-7 show the calculated changes in the unidirectional lamina properties as a function of change in the matrix modulus for various fiber volume fractions  $(V_f)$ .

Elast. Const.	Narmco 5208 Epoxy	T300 Carbon Fiber	Composite 5208/T300 .62V <sub>f</sub>
E <sub>11</sub>	.5	33.5	20.8
E22	.5	3.05	1.46
G <sub>12</sub>	.185	3.0	.633
<sup>v</sup> 12	.35	.285	.31

Table 1. Elastic Moduli of Narmco 5208/T300 graphite Fiber lamina and its constituents. (The values are given in 10<sup>6</sup> psi.)

## Example 2: Quick Estimate of the Ultimate Stiffness Change in a Composite.

We would like to know how long it takes for a 4-ply unidirectional T300/5208 composite to reach equilibrium in an environment such as the Panama Canal Zone, and what would be the expected percentage reduction of the elasticconstants of such a composite when tested at  $100^{\circ}C$  ( $212^{\circ}F$ ), as compared to the dry composite properties tested at the same temperature.

#### Known data:

Average temperature in the Canal Zone is about 77°F and the humidity if fairly constant about 93% RH.

The thickness of a ply of Narmco 5208/T300 after fabrication into a laminate is about .01397 cm.

The diffusion coefficient of the 5208/T300 composite at this temperature is about 1.0 x  $10^{-10}$  cm<sup>2</sup>/sec.

This problem is very simple and can be solved without further use of a computer.

From reference 8, Figure 4, we know that a 95% saturation value is given by  $Dt/h^2 = 1.25$  (where D is the diffusion coefficient, h is  $\frac{1}{2}$  the thickness of the laminate and t is the time to reach this percentage of equilibrium).

Since the thickness of a 4-ply laminate is .05588 cm, the time to reach 95% of the equilibrium concentration is about 16 weeks (see Figure 17 of reference 8).

Table 2. Calculated change of the lamina Young's modulus, E11, of a T300 composite as the matrix modulus  $E_m$  changes. (Values are given in 106 psi.)

EmVf	.4	.5	.6	.7
.5	13.70	17.00	20.30	23.60
.4	13.64	16.95	20.26	23.57
.3	13.58	16.90	20.22	23.54
.2	13.52	16.85	20.18	23.51
.1	13.46	16.80	20.14	23.48

Table 3. Calculated change of the lamina transverse Young's modulus, E22, of a T300 composite as the matrix modulus changes. (Values are given in 106 psi)

EmVf	.4	.5	.6	.7
.5	1.005	1.189	1.411	1.682
.4	.856	1.029	1.244	1.515
.3	.688	.844	1.042	1.304
.2	.496	.622	.790	1.023
.1	.271	.349	.459	.623

GmVf	.4	.5	.6	.7
.200	.415	.511	.642	.832
.175	.368	.455	.576	.753
.150	.320	.399	.506	.668
.125	.271	.338	.433	.577
.100	.220	.276	.356	.479
.075	.167	.211	.274	.374
.050	.113	.144	.188	.260
.025	.057	.073	.097	.135

Table 4. Calculated change of the lamina longitudinal shear modulus,  $G_{12}$ , of a T300 composite as the matrix shear modulus,  $G_m$ , changes. (Values are given in 106 psi.)

Table 5. Calculated change of the lamina transverse shear modulus, G23, of a T300 composite as the matrix shear modulus  $G_m$  changes. (Values are given in 106 psi.)

G <sub>m</sub> V <sub>f</sub>	.4	.5	.6	.7
.2	.882	.879	.946	1.085
.175	.809	.804	.867	1.000
.150	.730	.723	.781	.907
.125	.641	.634	.687	.803
.100	.543	.535	.581	.685
.075	.433	.425	.463	.552
.050	.308	.302	.330	.397
.025	.166	.161	.177	.216

Table 6. Calculated change in the lamina major Poisson's ratio,  $\nu_{12}$ , of a T300 composite as the matrix Poisson's ratio  $\nu_m$  changes. (Values are given in 106 psi.)

v <sub>m</sub> V <sub>f</sub>	.4	.5	.6	.7
.5	.414	.392	.371	.349
.4	.354	.342	.331	.319
.3	.294	.292	.291	.289
.2	.234	.242	.251	.259
.1	.174	.192	.211	.229

#### Table 7. Young's and Shear Moduli of Narmco 5208 Resin Equilibrated at Various Relative Humidities

Relative Humidity	Young's Mod. E <sub>m</sub> (PSIx10 <sup>6</sup> )		Shear Mod. G <sub>m</sub> (PSIx10 <sup>6</sup> )			Poisson's Ratio	
	22°C	100°C	22°C	100°C	22°C	100°C	
0	.5	.40	.185	.149	. 35	.35	
33		.34		.126		.35	
55		.31		.114	i	.35	
81		.26		.098		.35	
93		.21		.077		.35	

t.

From Figure 2 we see that at  $100^{\circ}$ C the retention in the matrix modulus is 81% of the room temperature value for the dry resin and only 41.5% for a resin equilibrated at 93% RH, i.e., if the room temperature resin modulus was .5 x  $10^{6}$  psi, the  $100^{\circ}$ C moduli for dry and wet resin are .405 x  $10^{6}$  and .207 x  $10^{6}$  psi.

From Figures 3-7 we can read off directly the changes in elastic constants of the composite. Assuming a fiber volume fraction of .6 we see from Figure 3 that no measurable change in E<sub>11</sub> is expected; from Figure 4 one can see that E<sub>22</sub> will drop from 1.46 x 10<sup>6</sup> to 1.25 x 10<sup>6</sup> and .8 x 10<sup>6</sup> psi respectively; from Figure 5 one can see the longitudinal shear modulus G<sub>12</sub> will drop from .633 x 10<sup>6</sup> psi to .512 x 10<sup>6</sup> and .263 x 10<sup>6</sup> psi respectively; from Figure 6 we see that the transverse shear modulus G<sub>23</sub> will drop from .90 x 10<sup>6</sup> to .729 x 10<sup>6</sup> and .373 x 10<sup>6</sup> psi respectively.

A comparison of the moduli of the dry and moisture exposed composites at 100 C shows that there is a 50% retention in the matrix modulus, no change in the tensile modulus  $E_{11}$ , a 64% retention in the transverse tensile modulus, a 51% retention of the longitudinal modulus  $G_{12}$  and a 51% retention of the transverse shear modulus  $G_{23}$ . Since it is not expected that the Poisson's ratio of the resin will change significantly below glass transition temperature, the change in the composite Poisson's ratio will be even less (see Figure 7).

The implications are, that not only are certain stiffness parameters reduced, but also the failure strength will be reduced if the strain to failure is not increasing correspondingly in order to permit the same state of internal stress.

At the present time we do not have enough information about the change in strain to failure of the matrix material as a function of temperature and absorbed moisture to compare experimental and calculated values, therefore, such considerations are still qualitative.

Example 3: The Temporal Change in Elastic Properties of a composite under Washington D. C. Weather Conditions.

An 8 ply unidirectional and an 8 ply  $\pm 45^{\circ}$  symmetric laminate (5208/T300; V<sub>f</sub> = .7) are thought to be exposed under Washington D. C. conditions. The properties of the matrix, fibers and dry composte are given in Table 1.

The internal distributions after 0.1, 2, 3, 5, 7, 8, 28, 45 and 50 months were calculated as indicated in reference 9. From Figure 2 (and Table 1 and 7) and by applying the  $\Delta$ -layer analysis the A, B and D matrices were calculated for the composite to be tested at  $100^{\circ}$ C. Only the A matrices for the exposure times of 0,3,8 and 45 months exposure are given in Table 8. Figure 8 shows the change in A<sub>66</sub> (the shear stiffness of the 8 ply unidirectional laminate). Table 8. "A Matrices" for 8-ply T300/Narmco 5208 (Vf = .7) laminates (0 = 0° and 0 =  $\pm 45^\circ$  symm.) after various

exposure times to Washington, D.C. weather conditions

the second s	Exposure Time (Months)	% Moisture	A Matrix Nonuniform Moist. Distr. (PSI)		£	A Matrix Uniform Moist. Distr. (PSI)	
	o	o	1.043E+6 2.043E+4 0 2.043E+4 6.709E+4 0 0. 0. 5	0. 0. 5.823E+4	1.043E+6 2.043E+4 0.	2.043E+4 6.709E+4 0.	0. 0. 5.823E+4
	m	.230	1.043E+6 1.958E+4 0 1.958E+4 6.431E+4 0 0. 0.	0. 0. 5.451E+4	1.043E+6 1.958E+4 0.	1.958E+4 6.430E+4 0.	0. 0. 5.443E+4
	œ	.881	1.042E+6 1.821E+4 0 1.821E+4 5.981E+4 0 0. 0. 4	0. 0. 4.870E+4	1.042E+6 1.825E+4 0.	1.825E+4 5.994E+4 0.	0. 0. 4.884E+4
	45	.919	1.041E+6 1.711E+4 0 1.711E+4 5.620E+4 0 0. 0. 4	0. 0. 4.434E+4	1.041E+6 1.711E+4 0.	1.711E+4 5.620E+4 0.	0. 0. 4.434E+4
	Ð	o	3.169E+5 2.587E+5 0 2.587E+5 3.169E+5 0 0. 0. 5	0. 0. 5.348E+5	3.169E+5 2.587E+5 0.	2.587E+5 3.169E+5 0.	0. 0. 5.348E+5
	m	.230	3.138E+5 2.593E+5 4 2.593E+5 3.138E+5 4 2.076E+2 2.076E+2 5	4.153E+2 4.153E+2 5.339E+5	3.137E+5 2.593E+5 0.	2.593E+5 3.137E+5 0.	0. 0. 5.339E+5
	œ	. 881	3.088E+5 2.601E+5 2 2.601E+5 3.008E+5 2 1.436E+2 1.436E+2 5	2.872E+2 2.872E+2 5.325E+5	3.089E+5 2.601E+5 0.	2.601E+5 3.089E+5 0.	0. 0. 5.325E+5
	45	919.	3.050E+5 2.606E+5 6 2.606E+5 3.050E+5 6 3.263E+1 3.263E+1 5	6.525E+1 6.525E+1 5.314E+5	3.050E+5 2.606E+5 0.	2.606E+5 3.050E+5 0.	0. 0. 5.314E+5

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NSWC/WOL TR 77-61

It also shows the change in strain the 8 ply  $\pm 45^{\circ}$  symmetric composite would show under a load of 500 pounds/inch in the 0° direction during this exposure (creep deformation excluded). One can see that these changes are proportional to the amount of moisture uptake.

Example 4: Effect of Internal Moisture Distribution.

It is of interest to answer the question of how important it is to know the internal moisture distribution in a composite in addition to the moisture content (the latter could be determined nondestructively). If there is an unsymmetric moisture distribution in a symmetric composite then the B matrix is no longer zero but if the moisture distribution is symmetric the B matrix remains zero.

We have chosen 6 cases:

Case 1: The same composite as given in the previous example.

Case 2 to Case 6 are shown in Figure 9 which show composites with arbitrary symmetric ply orientation and symmetric distribution of moisture, with unsymmetric ply orientation but symmetric moisture distribution, and with unsymmetric ply and moisture distributions. For the cases 2-6 a non linear  $E_m$  vs H<sub>2</sub>O concentration was assumed of the form  $C^2 + E^2 = 1$ . The Poisson's ratio of the matrix was assumed to remain constant (v = .35), the fiber properties are those of T300 and  $G_m = E_m / [2(1+v_m)]$ .

Table 8 shows the difference in the composite of case 1 with the actual and with a uniform moisture distribution of the same overall concentration. In this case there is no significant difference. The A matrices of the cases 2-6 are given in Table 9; also given are the percent differences in strains between the composites with uniform and nonuniform moisture distributions if a load of 500 lbs would be applied in the zero direction.

It is obvious that in general there is a difference in the elastic behavior between composites with nonuniform and uniform moisture distribution but with the same overall moisture content. It is not surprising that it shows up most strongly in simple +450 composites (case 3 and case 6) though, even there, with a "natural" distribution obtained under real conditions as in case 1, the % difference in strain (at a load of 500 lbs/inch) does not exceed 1% during the entire exposure time. Thus, in a first approximation one can say that the effect of the internal moisture distribution obtained under real conditions may be neglected compared to the stronger effect of the overall moisture content. Table 9. Effect of Internal Moisture Distribution on the Elastic Behavior of Composites.

<pre>% Difference in Strain ε<sub>x</sub> (at a Load of 500 lb/inch</pre>	٢	10	1.6		5.5
Distrib.	-9.581E+4	0	9.201E+4	9.139E+7	0
	9.581E+4	0	-9.383E+4	-9.321E+4	0
	4.887E+5	5.440E+5	3.521E+5	3.541E+5	5.448E+5
A Matrix	2.218E+5	2.495E+5	1.353E+5	1.532E+5	2.485E+5
Moisture	3.786E+5	3.509E+5	3.446E+5	3.469E+5	3.537E+5
(PSI)	4.790E+5	0	-4.692E+4	-4.661E+4	0
Uniform	3.786E+5	3.509E+5	5.492E+5	5.512E+5	3.537E+5
	2.218E+5	2.495E+5	1.535E+5	1.532E+5	2.485E+5
	-4.790E+4	0	4.600E+4	4.570E+4	0
Distrib.	-9.130E+4	4.584E+3	8.806E+4	9.197E+4	2.327E+3
	1.006E+5	4.584E+3	-9.327E+4	-9.326E+4	2.327E+3
	4.853E+5	5.407E+5	3.429E+5	3.475E+5	5.430E+5
A Matrix	2.226E+5	2.502E+5	1.514E+5	1.514E+5	2.491E+5
orm Moisture	3.676E+5	3.401E+5	3.378E+5	3.457E+5	3.477E+5
(PSI)	5.028E+4	2.292E+3	-4.663E+4	-4.663E+4	1.163E+3
A M	3.680E+5	3.401E+5	5.702E+5	5.453E+5	3.477E+5
Non-uniform Mo	2.226E+5	2.502E+5	1.514E+5	1.514E+5	2.491E+5
(P	-4.565E+4	2.292E+3	4.403E+4	4.599E+4	1.163E+3
Case	2	m	4	ŝ	9

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#### CONCLUSIONS

A compact formalism has been proposed to predict the changes in the elastic behavior of fiber reinforced composites by combining real outdoor weather data and a small number of laboratory measurements. The analysis is based on a combination of the theoretical concepts of diffusion, micromechanics and a modified laminated plate theory. It was also demonstrated that, while the overall moisture content in composites can change significantly the elastic properties, the changes of elastic properties due to the internal distribution obtained under natural conditions does not differ much from that of a uniform distribution with the same overall moisture content.

More work is required for predicting the change in composite strength as a result of moisture penetration.

#### ACKNOWLEDGEMENT

The authors would like to thank Messrs. M. Stander and C. Bersch of the Naval Air Systems Command for their interest and support of this work.



**4 PLY LAMINATE** 











FIG. 4 CHANGE OF COMPOSITE MODULUS (E22) AS A FUNCTION OF RESIN MODULUS IN T 300 CF UNIDIRECTIONAL COMPOSITES






G<sub>m</sub>, RESIN SHEAR MODULUS (10<sup>6</sup> PSI)















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FIG. 9 ARBITRARY MOISTURE DISTRIBUTION IN 8 PLY LAMINATES.

#### APPENDIX A

## PREDICTIONS OF THE ELASTIC CONSTANTS OF UNIDIRECTIONAL COMPOSITES FROM THE CONSTITUENT PROPERTIES

Longitudinal Modulus E<sub>11</sub> (= Young's Modulus)

The longitudinal modulus is usually described by the rule of mixtures

$$E_{11} = E_f V_f + E_m V_m \tag{A1}$$

or modified to include a fiber misalignment factor k

$$E_{11} = k (E_f V_f + E_m V_m)$$
(A2)

the rule of mixture was further modified by Ekval1[19]

$$E_{11} = E_f V_f + E'_m V_m \qquad E' = \frac{E_m}{1 - 2v_m^2}$$
 (A3)

where Ef is the longitudinal fiber modulus,  $E_m$  is the matrix modulus,  $V_f$  and  $V_m$  are the fiber and resin volume fractions and  $v_m$  is the resin Poisson's ratio.

Transverse Modulus E22

Haskin and Rosen derived the following relation [20, 21]

[19] J. C. Ekvall, AIAA 6th Structural and Materials Conf. Palm Springs, Apr 1965.

- [20] Z. Haskin and B. W. Rosen, "The Elastic Moduli of Fiber-
- Reinforced Materials." J. Appl. Mech. Jun 1964, p. 223.
- [21] B. W. Rosen, N. F. Don and Z. Haskin "Mechanical Properties of Fiber Composites." NASA CR-31 (1964).

$$E_{22} = E_{33} = \frac{4G_{23}K_{23}}{K_{23} - \psi G_{23}}$$
(A4)  
$$\psi = 1 + \frac{4K_{23} - \psi I_{2}^{2}}{E_{11}}$$

K23 and G23 are bulk and shear modulus governing plane-strain deformation in the 2,3-plane.

Tsai proposed [22]

$$E_{22} = 2 \left[ 1 - v_{f} + (v_{f} - v_{m}) V_{m} \right] \left[ (1 - C) \frac{K_{f} (2K_{m} + G_{m}) - G_{m} (K_{f} - K_{m}) V_{m}}{(2K_{m} + G_{m}) + 2 (K_{f} - K_{m}) V_{m}} + C \frac{K_{f} (2K_{m} + G_{f}) + G_{f} (K_{m} - K_{f}) V_{m}}{(2K_{m} + G_{f}) - 2 (K_{m} - K_{f}) V_{m}} \right]$$
(A5)

where

$$\begin{split} \kappa_{f} &= E_{f}/2(1-\nu_{f}) & \nu_{f} &= \text{ fiber Poisson's ratio} \\ \kappa_{m} &= E_{m}/2(1-\nu_{m}) & \nu_{m} &= \text{ matrix Poisson's ratio} \\ G_{f} &= E_{f}/2(1+\nu_{f}) & c &= o \text{ for isolated fibers} \\ G_{m} &= E_{m}/2(1+\nu_{m}) & c &= 1 \text{ for fibers in contact} \end{split}$$

or approximated by

$$E_{22} = \frac{E_f E_m}{E_m V_f + E_f V_m}$$
(A6)

Major Poisson's ratio v<sub>12</sub>

$$v_{12} = v_f V_f + v_m V_m \qquad (= rule of mixture) \qquad (A7)$$

[22] S. W. Tsai, "Structural Behavior of Composite Materials," NASA CR-71 (1964).

A refinement by Rosen [20]

$$v_{12} = \frac{E_{f}V_{f}L_{1} + V_{m}E_{m}L_{2}v_{m}}{V_{f}E_{f}L_{3} + V_{m}E_{m}L_{2}}$$

$$L_{1} = 2v_{f}(1-v_{m}^{2})V_{f} + V_{m}(1-v_{m})v_{m}$$

$$L_{2} = V_{f}(1-v_{f}-2v_{f}^{2})$$

$$L_{3} = 2(1-v_{m}^{2})V_{f} + (1+v_{m})V_{m}$$

Tsai proposed (A9) [22]  

$$v_{12} = (1-C) \frac{K_{f}v_{f}(2K_{m}-G_{m})V_{f}+K_{m}v_{m}(2K_{f}+G_{m})V_{m}}{K_{f}(2K_{m}+G_{m})-G_{m}(K_{f}-K_{m})V_{m}}$$

+ C 
$$\frac{K_{m}v_{m}(2K_{f}+G_{f})V_{m}+K_{f}v_{f}(2K_{m}+G_{f})V_{f}}{K_{f}(2K_{m}+G_{f})+G_{f}(K_{m}-K_{f})V_{m}}$$
(A9)

(A8)

where the symbols are the same as defined under (A5). Longitudinal Shear Modulus,  $G_{12}$ 

Ekvall [23] proposed (Al0) and (All)

$$G_{12} = \frac{G_f G_m}{V_m G_f + V_f G_m}$$
 (for square filaments) (A10)

$$G_{12} = \frac{G_{f}G_{m}}{R\phi G_{m} + (1-R)G_{f}}$$
 (for round filaments) (All)

where

$$\phi = \int_{O}^{2\pi} \frac{\sin \theta d\theta}{(G_m/G_f) + \sin \theta 1 - (G_m/G_f)}$$

$$R = \frac{4V_{f}}{\pi (V_{f} + V_{m})} = \frac{4V_{f}}{\pi} = \frac{4(1 - V_{m})}{\pi}$$

[23] J. C. Ekvall, ASME paper 61 AV-56, Aviation Conf., Los Angles, Mar 1961.

Rosen [20] proposed an upper (A10) and a lower bound (A11) for  $G_{12}$ 

$$G_{12} \text{ (upper bound)} = G_m (m_G V_1 + V_2) \tag{A12}$$

$$G_{12}$$
 (lower bound) =  $G_m (V_1/m_G) + V_2$  (A13)

where

$$v_{1} = \frac{\pi}{2\sqrt{3}} \qquad v_{2} = 1 - v_{1}$$

$$m_{G} = \frac{n(1 + \beta^{2}) + (1 - \beta^{2})}{n(1 - \beta^{2}) + (1 - \beta^{2})}$$

$$\eta = \frac{G_{\varepsilon}}{G_{m}} \qquad \beta = \frac{v_{f}}{v_{1}}$$

for a random array he proposed

$$G_{12} = G_{m} \frac{\eta (1+V_{f}) + V_{m}}{\eta V_{m} + 1 + V_{f}}$$
(A14)

Tsai [22] proposed

$$G_{12} = (1-C)G_{m} \frac{2G_{f} - (G_{f} - G_{m})V_{m}}{2G_{m} + (G_{f} - G_{m})V_{m}} + CG_{f} \frac{(G_{f} + G_{m}) - (G_{f} - G_{m})V_{m}}{(G_{f} + G_{m}) + (G_{f} - G_{m})V_{m}}$$
(A15)

Transverse Shear Modulus G<sub>23</sub> Rosen proposed **[**20, 21**]** 

$$G_{23} = G_{m} \frac{\left[ (\alpha + \beta_{m} V_{f}) (1 + \rho V_{f}^{3}) - 3V_{f} (1 - V_{f}) \beta_{m}^{2} \right]}{\left[ (\alpha - V_{f}) (1 + \rho V_{f}^{3}) - 3V_{f} (1 - V_{f})^{2} \beta_{m}^{2} \right]}$$
(A16)

where

$$\alpha = \frac{\gamma + \beta_{m}}{\gamma - 1}, \quad \beta_{m} = \frac{1}{3 - 4\overline{\nu}_{m}}, \quad \beta_{f} = \frac{1}{3 - 4\overline{\nu}_{f}}$$

$$\rho = \frac{\beta_{m}}{1 + \gamma\beta_{f}} \frac{-\gamma\beta_{f}}{f}, \quad \gamma = \frac{G_{F}}{G_{m}} \qquad G_{F} = G_{F_{12}} = G_{F_{23}}$$

$$A - 4$$

# Halpin-Tsai Equations.

For design purposes and rapid computational procedures Halpin and Tsai 11 have proposed a set of equations which are simpler than most of the above formulas but are good enough for estimating with reasonable accuracy the ply properties:

$$E_{11} \simeq E_f V_f + E_m V_m$$
(A17)

$$v_{12} \approx v_f V_f + v_m V_m$$
(A18)

$$\frac{\overline{P}}{P_{m}} = \frac{(1+\zeta_{n}V_{f})}{(1-\gamma V_{f})}$$
(A19)

where

$$n = [(p_f/p_m) - 1] / [(p_f/p_m) + \zeta]$$

 $\overline{p}$  = composite moduli  $E_{22}$ ,  $G_{12}$ , or  $G_{23}$ ;  $p_f$  = corresponding fiber modulus  $E_{11f}$   $G_{12f}$   $v_{12f}$  (or  $v_{23f}$ );  $p_m$  = corresponding matrix modulus  $E_m$ ,  $G_m$ ,  $v_m$ ;  $\zeta$  = a measure of reinforcement which depends on the boundary conditions. Approximate values are given by  $\zeta_{E_{22}}$  = 2 (a/b) and by log  $\zeta G_{12}$  = 3 log (a/b), i.e., for round fibers  $\zeta_{E_{22}}$  = 2 and  $\zeta_{G_{12}}$ 

# APPENDIX B

Useful relationships between A, B and D matices.

From

$$\begin{bmatrix} -\frac{N}{M} \end{bmatrix} = \begin{bmatrix} -\frac{A}{B} - \frac{1}{D} - \frac{B}{D} \end{bmatrix} \begin{bmatrix} \varepsilon^{O} \\ \kappa \end{bmatrix}$$
(B1)

follows

$$\begin{bmatrix} N \end{bmatrix} = \begin{bmatrix} A \end{bmatrix} \begin{bmatrix} \varepsilon^{\circ} \end{bmatrix} + \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} \varepsilon \end{bmatrix}$$
 (B2)   
 
$$\begin{bmatrix} \varepsilon^{\circ} \end{bmatrix} = \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} N \end{bmatrix} - \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} \kappa \end{bmatrix}$$

$$\begin{bmatrix} M \end{bmatrix} = \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} N \end{bmatrix} - \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} K \end{bmatrix}$$

$$\begin{bmatrix} M \end{bmatrix} = \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} N \end{bmatrix} + (\begin{bmatrix} D \end{bmatrix} - \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix}) \begin{bmatrix} K \end{bmatrix}$$
(B3)

$$\begin{bmatrix} \varepsilon^{\circ} \\ -W^{-} \end{bmatrix} = \begin{bmatrix} A^{*} \\ -\overline{C} \\ + \overline{C} \end{bmatrix} \begin{bmatrix} -N \\ -W^{-} \end{bmatrix} \begin{bmatrix} -N \\ -W^{-} \end{bmatrix}$$
(B4)

$$\begin{bmatrix} M \end{bmatrix} \begin{bmatrix} C^* \end{bmatrix} D^* \end{bmatrix} \begin{bmatrix} K \end{bmatrix}$$
$$\begin{bmatrix} A^* \end{bmatrix} = \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix}$$
$$\begin{bmatrix} C^* \end{bmatrix} = \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix}$$
$$\begin{bmatrix} D^* \end{bmatrix} = \begin{bmatrix} D \end{bmatrix} - \begin{bmatrix} B \end{bmatrix} \begin{bmatrix} A \end{bmatrix}^{-1} \begin{bmatrix} B \end{bmatrix}$$

B-1

#### APPENDIX C

#### PROGRAM TO CALCULATE LAMINATE MATERIAL PROPERTIES

Below we outline a computer program which calculates the time history of the elastic properties of a laminate comprised of transversely isotropic unidirectional plys of a carbon fiber reinforced epoxy composite when both faces of the laminate are exposed to prescribed climatic conditions.

A program is already available which calculates the time dependent moisture distribution within such a laminate [3, 9]. The computer program described in this section calculates the elastic properties in such a laminate when given; its geometry, its internal moisture distribution, its temperature, the dependence of the elastic properties of the fiber and of the resin on internal moisture concentration and temperature, and the dependence of the elastic properties of a ply of the composite upon the elastic properties of the fiber and of the resin.

#### NOTATION

The laminate lies in the region  $-\overline{z} \le z \le \overline{z}$ . This region is subdivided into  $\Delta$ -layers whose endpoints  $P_k = P(k)$  are located at

 $-\overline{z} = P(1) < P(2) < \dots < P(NDL+1) = \overline{z}$ 

where NDL stands for number of delta layers. These points will be symmetrically located about 0 = P(1 + NDL/2), and each interval [P(k), P(k+1)] will lie within one ply of the laminate.

The values of internal moisture concentration (at any given time t) at the points P(k) determined using the program described in [9] are denoted by C(k) (k=1, ..., NDL+1). The points P(k) are chosen to be spaced closely enough so that, as far as determining the material properties within each delta layer  $\Delta_k = [P(k), P(k+1)]$  the moisture concentration within  $\Delta_k$  can be taken to have the constant value W = (C(k) + C(k+1)/2. Assuming that the elastic properties of each delta layer can be calculated given W and the temperature, the problem of determining the elastic properties of the laminate is reduced to classical laminated plate theory, as described earlier in this text.

# FORMULAS USED FOR LAMINA ELASTIC CONSTANTS

The matrix elastic constants  $E_m$  and  $v_m$  (or  $G_m = E_m/(2(1+v_m))$ are assumed to be given by formulas or tables as functions of uniform internal moisture concentration and temperature, and the fiber elastic constants EllF, Gl2F,  $v_{12F}$  are taken to remain constant. For predicting the lamina properties Ell, E22, Gl2, and  $v_{12}$ , the Halpin-Tsai equations (Al7-Al9) were used (here the 1-direction is parallel to the fibers);

$$E_{11} = E_{11f} V_f + E_m V_m \tag{C1}$$

$$v_{12} = v_{12f} v_f + v_m v_m$$
 (C2)

where  $V_f$  = fiber volume fraction and  $V_m = 1 - V_f$  = matrix volume fraction. Also

$$\overline{P} = \frac{(1 + \zeta v V_f)}{(1 - \eta V_f)} P_m$$
(C3)

where

 $v = \left[ (P_f/P_m) - 1 \right] / \left[ (P_f/P_m) + \zeta \right]$ 

and where  $\overline{P}$  is either E<sub>22</sub> or G<sub>12</sub> with P<sub>f</sub> the corresponding fiber modulus (E<sub>22f</sub> or G<sub>12f</sub>) and with P<sub>m</sub> the corresponding matrix modulus (E<sub>m</sub> or G<sub>m</sub>). For round fibers the parameter  $\zeta$  is given by  $\zeta$ (E<sub>22</sub>) = 2 and  $\zeta$ (G<sub>12</sub>) = 1. For calculating G<sub>23</sub>, Rosen's equation (A16) was used;

$$G_{23} = G_{m} \frac{(\alpha + \beta_{m} V_{f}) (1 + \rho V_{f}^{3}) - 3V_{f} (1 - V_{f}) \beta_{m}^{2}}{(\alpha - V_{f}) (1 + \rho V_{f}^{3}) - 3V_{f} (1 - V_{f}) \beta_{m}^{2}}$$
(C4)

where

$$\alpha = \frac{\gamma + \beta_m}{\gamma - 1} , \beta_m = \frac{1}{3 - 4\nu_m}, \beta_f = \frac{1}{3 - 4\nu_f}, \rho = \frac{\beta_m - \gamma \beta_f}{1 + \gamma \beta_f}, \gamma = \frac{G_{12f}}{G_m}$$

We now give the notation used in the computer program for the quantities in equations (10), (12), (15), (16), (27), and (28). The angle of the k<sup>th</sup> delta layer (the counterclockwise angle in degrees through which the positive x-axis must be rotated to reach the positive 1-axis) is denoted by  $\theta(k)$ . For each k; Q11, Q12, Q22, Q44, Q55, Q66 are given by (3)-(5);

$$Q_{11} = \frac{E_{11}}{(1-v_{12}v_{21})}, \ Q_{22} = \frac{E_{22}}{(1-v_{12}v_{21})}, \ Q_{44} = G_{23}, \ Q_{55} = Q_{66} = G_{12},$$

$$Q_{12} = Q_{21} = \frac{v_{21} E_{11}}{(1 - v_{12} v_{21})} = \frac{v_{12} E_{22}}{(1 - v_{12} v_{21})} \cdot$$

Define  $m = \cos(\theta(k))$  and  $n = \sin(\theta(k))$  and define [10], [12];

Q11 BAR(k) = 
$$Q_{11} m^4 + 2(Q_{12}+2Q_{66}) m^2 n^2 + Q_{22} n^4$$
  
Q22 BAR(k) =  $Q_{11} n^4 + 2(Q_{12}+2Q_{66}) m^2 n^2 + Q_{22} m^4$   
Q12 BAR(k) =  $(Q_{11}+Q_{22}-4Q_{66}) n^2 m^2 + Q_{12} (m^4 + n^4)$   
Q66 BAR(k) =  $(Q_{11}+Q_{22}-2Q_{12}-2Q_{66}) m^2 n^2 + Q_{66} (m^4 + n^4)$  (C5)  
Q16 BAR(k) =  $(Q_{11}-Q_{12}-2Q_{66}) nm^3 + (Q_{12}-Q_{22}+2Q_{66}) n^3 m$   
Q26 BAR(k) =  $(Q_{11}-Q_{12}-2Q_{66}) n^3 m + (Q_{12}-Q_{22}+2Q_{66}) nm^3$   
Q44 BAR(k) = Q44 m^2 + Q55n^2  
Q55 BAR(k) = Q44 n^2 + Q55m^2  
Q45 BAR(k) = (Q55-Q44) mn  
For i, j = 1,2,6 define:

$$A_{ij} = \sum_{k=1}^{NDL} Q_{ij} BAR(k) \cdot (P(k+1) - P(k))$$

$$B_{ij} = \sum_{k=1}^{NDL} Q_{ij} BAR(k) \cdot (P(k+1)^2 - P(k)^2)/2$$
(C7)

$$D_{ij} = \sum_{k=1}^{NDL} Q_{ij} BAR(k) \cdot (P(k+1)^3 - P(k)^3)/3$$
(C8)

and for i, j = 4,5 (and  $ATS_{ij}$  the coefficients for transverse shear)

$$ATS_{ij} = 1.25 \sum_{k=1}^{NDL} Q_{ij} BAR(k) P(k+1) - P(k) - \frac{4(P(k+1)^3 - P(k)^3)}{3(2\overline{z})^2}$$

The 3 by 3 matrix A is defined to have the entries

	/A11	A12	2A16
A =	A12	A22	2A26
	A16	A26	2A66/

and similarly with B and D.

The 3 by 3 matrices ASTAR, BSTAR, CSTAR, DSTAR are given (in terms of matrix algebra) by (Appendix B)

ASTAR =  $A^{-1}$ BSTAR =  $-A^{-1}B$ CSTAR =  $BA^{-1}$ DSTAR =  $D-BA^{-1}B$ .

(C9)

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