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## MACROMOLECULAR DIVISION
## COMMISSION ON POLYMER CHARACTERIZATION AND PROPERTIES
## WORKING PARTY ON STRUCTURES AND PROPERTIES OF COMMERCIAL POLYMERS*

### Intrinsic Characterization of Continuous Fibre Reinforced Thermoplastic Composites—II

# PSEUDO-ELASTIC CONSTANTS FOR AROMATIC POLYMER COMPOSITE (APC-2)

(Technical Report)

*Prepared for publication by*

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# Intrinsic characterization of continuous fibre thermoplastic composites—II. Pseudo-elastic constants for aromatic polymer composite (APC-2) (Technical Report)

ABSTRACT: Tensile characteristics, stiffness and ultimate strength of laminates of different constructions, namely uniaxial, cross- and angle-plied, are measured and compared with the theoretical values generated by means of the micro- and macromechanical modelling. Stiffness characteristics, including the engineering constants of the CF/PEEK lamina, have been found to be predictable from first principles, i.e. from the properties of the constituting materials and their relative concentrations. The ultimate strength can be derived providing that the laminate response is linear and elastic. This is not the case for angle-plied laminates where ply-reorientation complicates the prediction.

## 1. INTRODUCTION

IUPAC Working Party 4.2.1 (Structure and Properties of Commercial Polymers) has been involved in a series of studies of the mechanical and morphological behaviour of continuous fibre reinforced thermoplastic composites. The paper by Moore and Seferis (ref.1) addresses the toughness aspects. The aim of this paper is to address the engineering (pseudoelastic) constants that characterise the material under study - Aromatic polymer composite APC-2 of ICI based on 62% by volume of carbon fibres (AS4) impregnated with a polyether etherketone (PEEK) matrix

The following six laboratories participated in this study:

I) Polymeric Composites Laboratory, University of Washington, Seattle, USA (J.C. Seferis)
II) Huls AG, Marl, FRG (H. Schwickert)
III) ICI plc, Materials Centre, Wilton, UK (D.R. Moore)
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The aims of the work have been several fold:

(a) to characterise experimentally the fundamental building block - the lamina - by measuring the engineering properties (defined in the theoretical part) of uniaxial laminates;
(b) to calculate these engineering properties from those of constituents and their relative concentrations;
(c) to check the validity of the micromechanical modelling by confronting the information from a) with that from b);
(d) to measure the stiffness and the failure characteristics - ultimate strength and strain - of laminates of more complex constructions;
(e) to predict laminate properties by means of the Classical Lamination Theory (CLT);
(f) to correlate experimental data from d) with theoretical predictions from e) and to assess the potential of the macromolecular model.

## 2. THEORETICAL

The goal is to outline a model that allows the two fundamental properties - stiffness and ultimate strength - for any laminate construction to be derived on both the micro- and macromechanical level of abstraction. In this context, the following definitions are appropriate:

(a) Lamina and the uniaxial laminate are synonyms; (b) Stiffness is the operator (a matrix for anisotropic materials such as composites) relating the stress and strain tensors; (c) The ultimate strength is the stress level causing total failure; (d) The uniaxial laminate satisfies the conditions of orthotropic anisotropy under the plane stress situation, i.e. the properties are the same in all directions perpendicular to the fibres but different from those associated with the direction parallel to the fibres and its thickness is negligible with dimensions in the laminate plane; (e) Micromechanical abstraction considers the composite to be heterogeneous and provides a means of predicting lamina performance on the basis of the material properties and relative concentrations of the constituent (matrix and fibre) phases; (f) the macromechanical concept considers the composite to be homogeneous, with the constituents' effects detected only as average effective properties allowing experimental data measured on test coupons to be utilised in the design analysis of more complicated structures.

The stress/strain $\sigma/\epsilon$ behaviour of any composite can be written as:

$$\{\sigma\} = |Q|\{\epsilon\} \qquad (1)$$

*(see Note a)*

where $|Q|$ stands for the stiffness matrix. For an orthotropic lamina under plane stress situation and loaded in one of the principal directions, only four matrix elements are required:

$$|Q| = \begin{vmatrix} Q_{11} & Q_{12} & 0 \\ & Q_{22} & 0 \\ & & Q_{66} \end{vmatrix} \qquad (2)$$

with the $Q_{ij}$ elements readily relatable to the Engineering constants. These are the longitudinal modulus $E_1$ associated with the property along the reinforcement, the transverse modulus $E_2$ related to the properties perpendicular to the reinforcement, the shear modulus $G_{12}$and the Poisson ratio $\mu_{12}$ both measured in the lamina plane.

The lamina concept is very powerful in composite science as it allows the Engineering constants 1) to be measured and thus to enumerate the four matrix stiffness elements, 2) to be predicted micromechanically and 3) to be related to laminate properties regardless of the laminate construction and loading direction. The following two subsections deal with points 2) and 3).

### 2.1. Micromechanics

Under the usual assumptions (ref. 2) concerning spacing of the parallel reinforcement, linear elastic behaviour of the matrix, isotropic nature of the constituents and equality between adhesive and cohesive properties of the matrix, the two principal moduli can be derived from the simple rules of mixtures:

$$E_1 = E_f v_f + E_m(1-v_f) \qquad (3)$$

$$1/E_2 = v_f/E_f + (1-v_f)/E_m \qquad (4)$$

with $E_f$ and $E_m$ being the elastic moduli of the reinforcing fibre and the matrix respectively and $v_f$ the volume fraction of the reinforcement.

Whereas the Poisson ratio $\mu_{12}$ is given by an equation similar to (3), the shear modulus $G_{12}$ can be derived using the Halpin-Tsai semi-empirical approach (ref. 3):

$$G_{12} = G_m\ (1 + \xi\lambda\ v_f)/(1 - \lambda v_f) \qquad (5)$$

with $\xi$ and $\lambda$ being functions of the ratio of shear moduli $G_f/G_m$ and derived (ref. 4) as

$$\xi = (1 + G_m/G_f)/2 \qquad \lambda = (G_f/G_m-1)/(G_f/G_m+ \xi) \qquad (6)$$

As the algorithms for predicting the lamina strength in its principal directions are still rather underdeveloped, no attempt is made to derive theoretically the longitudinal, transverse and shear stress characteristics X, Y and S.

### 2.2. Macromechanics

Unlike the case of orthotropic lamina loaded in one of the principal directions, off-axis stressing or a more complex laminate construction cause coupling between shear strain and normal stresses and between shear stress and normal strains. Whilst

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Note a: The IUPAC recommendations to type the symbols for physical quantities in italic has not been followed in this manuscript; re-typing could not be justified on account of cost and publication delays.

the laminate performance is still characterised by the four 'Material characteristics' typifying the lamina, the theoretical approach requires also information on the laminate construction, namely the number of plies n and fibre orientation $\alpha_i$ within each ply with respect to a fixed coordinate system and the direction $\phi$ of load applied during testing in relation to the same coordinate system.

The model itself constitutes the "Classical Lamination Theory" utilising the mathematical expressions contained e.g. in refs. 4-7. The following is a brief outline of operations involved; more details are given in ref. 8.

The theoretical stiffness is derived in four steps: i) Conversion of the Engineering constants into elements of the stiffness matrix $|Q|$ using the equations 1 and 2, ii) Transformation of $|Q|$ into the transformed reduced stiffness matrix $|\overline{Q}|$ employing the transformation matrix $|T|$, iii) calculation of the extensional matrix $|A|$ by means of the ply index concept (ref. 4) and iv) extraction of laminate stiffness $E(\phi) = 1/a_{11}$ being the appropriate element of the inverse matrix $|a|=|A|^{-1}$. The transverse and shear moduli are readily obtainable as $E(\phi+90)=1/a_{22}$ and $G(\phi)=1/a_{33}$.

The ultimate laminate stiffness is calculated in five additional steps: v) derivation of the dimensionless 'Force distribution matrix' $|f|$ that relates the stress level within given a ply to the exerted force N for a symmetrical laminate under a simple deformation regime (uniaxial tension in our case), vi) the calculation of force $N_i$ acting on an individual ply i from the Tsai-Hill failure criterion (ref. 9), vii) determination of the stress/strain coordinates of the "knee-point" that corresponds to the first ply failure, viii) elimination of all failed lamina f by redefinition $Q_{12}(f) = Q_{22}(f) = Q_{66}(f) = 0$, $Q_{11}(f) = Q_{11}(i)$ and repeating the operations 2-6 with the goal to characterise the post-knee regime in terms of the ultimate stiffness. The final step ix) involves calculation of the ultimate strength $\sigma(\phi)$ by analysing the balance for the terminal plies.

The Tsai-Hill failure criterion is based on:

$$\sigma_1{}^2/X^2 - \sigma_1\sigma_2/X^2 + \sigma_2^2 + Y^2 + \tau_{12}{}^2/S^2 = 1$$

with $\sigma_1$ and $\sigma_2$ being the stresses calculated to have developed in the longitudinal and transverse directions and $\tau_{12}$ that in shear and in the laminate plane.

## 3. EXPERIMENTAL

### 3.1. Materials

Two laminate constructions have been investigated: uniaxial and cross-plied. The uniaxial laminates were made available in two forms: 1) as $[0]_{16}$ prepared by means of autoclaving and $[0]_8$ by compression moulding. The cross-plied construction was prepared in one thickness only of 8 plies regardless of the manufacturing route. The stacking sequence conformed to that usually termed as regular symmetrical, i.e $[0/90/0/90]_S$. The symmetrical construction has been chosen to decouple extension from bending which might have been an unnecessary complication both from the experimental and theoretical points of view. Both manufacturing techniques seem to give materials identical in their lamina properties.

The experimental conditions for manufacturing in the autoclave were those described in ref. 1. The compression moulding was done in a picture-frame mould between two 400x400x10 mm aluminium platens. Stacking was done parallel to the frame edges. Any undesirable movement of the APC-2 precuts was prevented by point-welding by means of soldering iron. Aluminium sheets of 0.2 mm protected the platten surfaces against possible PEEK contamination and very thin Al cooking foil was used instead of a chemical agent for quick release. The moulding operation was carried out in three steps on two Bucher presses (Bucher-Guyer AG, Niederweningen, Switzerland). The smaller, 35 tonnes machine was used first to "contact" the plies for 10 min at 380°C under a force of 68 kN and then to "consolidate" the laminate at the same temperature for 15 min under a force of 195 kN. After that the mould was transferred (typically 70-80 secs) into a larger (100 tonne) press operating at 190°C in which the laminates were cooled for 15 min under a load of 274 kN. Stress build-up was minimised by quick release of the laminate from the picture frame, trimming the edges and removing the thin Al foil.

### 3.2. Testing

The number of experimental approaches was proportional to the number of the involved laboratories. With the exception of the testing temperature (23°C) and the way of cutting test coupons (water cooled diamond saw) the methods used for obtaining the tensile properties differed in: a) testing machines (Instron testing machines 8033 servohydraulic, 1195 universal, the Zwick 1322 tensile tester with an installed liquid dilatometer system), b) standard procedures (ref. 10, 11) followed, c) strain rates ($1.7*10^{-4}$ - $7.2*10^{-4}$ $sec^{-1}$), d) tabbing (tabs used or replaced by gripping between two sheets of Metalit K247, grade 100 sandpaper), e) coupon and gauge length (the latter between 75 and 130 mm), f) monitoring the state of strain (optical, clip-on 25 mm extensometer) and, finally, g) number of measurements from which average properties have been derived.

Whilst no experimental difficulties have been encountered during testing the uniaxial laminates, studies of more complex constructions were associated with some problems. For the cross-plied laminates, we found it important to test them with the stiffer ply (fibre direction coincides with loading) in the top and bottom laminate surfaces. The alternative approach with the softer (90°) plies in direct contact with the grips usually resulted in lower modulus values. For the +/- 45° angle-plied laminates, a highly non-linear $\sigma/\epsilon$ response has been observed from which determination of the initial modulus was inaccurate. Thus a graphical technique using convergence of tangential and secant moduli, both plotted against the square root of stress, was developed and the initial material modulus identified from the intercept of the two lines.

Two different procedures have been followed to determine the shear modulus $G_{12}$: i) by testing at the angle of 45° and calculating $G_{12}$ as (ref. 12):

$$1/G_{12} = 4/E(45) - 1/E_1 - 1/E_2$$

neglecting the term $2\mu_{12}/E_1$ and ii) by conducting torsion experiments on both the longitudinal specimens (shear in the plane 1-2) and the transverse coupons (shear in the plane 1-3) following the procedure described before (ref. 13).

## 4. RESULTS AND DISCUSSION

### 4.1. Lamina behaviour – micromechanics

Table I is a compendium of all tests carried out at 23°C with the uniaxial laminates. Moreover, it records the manufacturing routes used, reports data as given by the manufacturer (ref. 14) and gives the data calculated by means of the micromechanics.

Table I: Tensile characteristics of the lamina

| LABORATORY MANUFACTURING | III Autoclave | IV Compression moulding | DATA SHEET | THEORY |
|---|---|---|---|---|
| $E_1$ [GPa] | 130 | 135.2 +/- 8.5 | 134 | 144 |
| $E_2$ [GPa] | 9.7 | 9.2 +/- 0.4 | 8.9 | 9.9 |
| $G_{12}$ [GPa] | | 4.9 +/- 0.5 | 5.1 | 4.5 |
| $G_{13}$ [GPa] | 5.5 | | | |
| $G_{23}$ [GPa] | 4.4 | | | |
| $\mu_{12}$ | 0.35 | | | 0.34 |
| $\mu_{13}$ | 0.30 | | | |
| $\mu_{21}$ | 0.04 | | | |
| X [MPa] | | 2090 +/- 80 | 2130 | |
| Y [MPa] | | 73 +/- 9 | 80 | |
| S [MPa] | | 78 | | |
| $\epsilon_1$ [%] | | 1.43 +/- 0.01 | 1.45 | |
| $\epsilon_2$ [%] | | 0.88 +/- 0.16 | 1.0 | |

Invariance in the experimental data with respect to the manufacturing route and a very good agreement with the manufacturer's specification indicate APC-2 to be a robust and tolerant system lending itself to easy characterisation. The following lamina characteristics are proposed to characterise the APC-2 system:

| Moduli [GPa] Normal | Shear | Strength [MPa] Normal | Shear | Poisson ratios in-plane | out-plane | Elongation [%] |
|---|---|---|---|---|---|---|
| $E_1$=132.6 | $G_{12}$=4.9 | X=2090 | S=78 | $\mu_{12}$=0.35 | $\mu_{13}$=0.30 | $\epsilon_1$=1.43 |
| $E_2$= 9.45 | $G_{13}$=5.5 | Y= 73 | | $\mu_{21}$=0.04 | | $\epsilon_2$=0.88 |
| | $G_{23}$=4.4 | | | | | |

As the underlined characteristics will be used as the input for the macromechanical analysis, steps have been taken to relate them to the values calculated by means of the micromechanical modelling. All the micromechanical calculations were carried out with behaviour of other polymers - ref.17), of the fibre the Table I. The relative errors with respect to the lamina characteristics quoted above appear to be as:

$E_1$ = -7.9%  $E_2$ = -4.5%  $G_{12}$ = +8.9%  $\mu_{12}$ = +2.9%

The fact that the error never exceeds 10% allows us to draw a very positive conclusion, namely that the micromechanical prediction of the APC-2 lamina properties is possible in spite of a very simple model having been used. Furthermore, the observed consistency in the experimental data combined with the theory/experiment fit vindicates the underlined lamina characteristics as being meaningful inputs for the macromechanical modelling of more complex laminates.

### 4.2. Laminate behaviour – macromechanics

Whilst the cross-plied construction has been characterised by one laboratory only with the results given in Table II, the +/- 45° angle-plied construction enjoyed involvement of four laboratories. The results of their measurements are given in Table III.

**Table II: Tensile characteristics of the cross-plied laminates**

| LABORATORY MANUFACTURING ROUTE | | III compr.mould. | | THEORY |
|---|---|---|---|---|
| | | mean | st.dev | |
| INITIAL STIFFNESS | [GPa] | 71.3 | 3.6 | 71.5 |
| FIRST-PLY FAILURE: STRESS | [MPa] | | | 556 |
| TERMINAL STIFFNESS | [GPa] | | | 66.8 |
| ULTIMATE STRENGTH | [MPa] | 1035 | 26 | 1163 |
| ULTIMATE STRAIN | [%] | 1.65 | 0.03 | |

**Table III: Tensile characteristics of the angle-plied laminates**

| LABORATORY MANUFACTURING | | II AUTOCL | IV COMPR.MLD | V AUTOCL | VI AUTOCL | THEORY |
|---|---|---|---|---|---|---|
| STIFFNESS | [GPa] | 10.2 | 16.6 | 19.1 | 16.7 | 17.3 |
| st.dev. | | 1.06 | 1.0 | | 1.3 | |
| ULTIMATE STRESS | [MPa | 264 | 253 | 286 | 314 | 153 |
| st.dev. | | 20 | 29 | | 8 | |
| ULTIMATE STRAIN | [%] | 13.5 | 19.2 | 9.6 | 18.3 | |
| st.dev. | | 1.0 | 1.9 | | 1.0 | |

The theoretical analysis has been carried out with the lamina characteristics (underlined values stated in the Table in Chapter 4.1) reflecting the material properties stemming from the PEEK/carbon fibre partnership and details of the laminate constructions. These were the number of plies n=8, fibre orientations in individual plies $\alpha_1 = \alpha_3 = \alpha_6 = \alpha_8 = 0$, $\alpha_2 = \alpha_4 = \alpha_5 = \alpha_7 = 90°$ and the loading direction $\phi$ being either 0° or 45°.

For the cross-plied laminates, experimentally determined values of the initial stiffness and the ultimate strength agree well with the theoretical predictions. The relative errors normalised on the appropriate theoretical value are -0.3% for the stiffness and -11% for the ultimate strength. The experimental stress/strain response has been measured as linear in line with the theory which predicts the first-ply failure of the transverse plies to be responsible only for 6.6% reduction in the laminate stiffness.

For the angle-plied laminates, the situation is more complex. Firstly, the experimental information on the laminate stiffness spans an alarmingly wide band of 10.2 to 19.1 GPa with three laboratories relatively close in their measurements and the laboratory quoting the lowest value more than 60% out. If one may disregard this low value, then the average experimental stiffness is calculated as 17.5 +/- 1.4 GPa, in satisfactory agreement with the theoretical value. The relative error is derived as +1.1% on the prediction. Secondly, four sets of data on the ultimate strength lay within a relatively narrow band of 279 +/- 27 MPa, however the mean is much higher - more than 80% - than the calculated value. Additional studies outside the IUPAC programme (ref. 19) revealed that a mechanism of ply-angle closing is responsible for this discrepancy. The ply-closing happens because the fibres embedded in the thermoplastic matrix adjust their orientation on composite extension. Typically, the original +/- 45° construction fails as +/- $\alpha$ with the terminal ply-angle $\alpha$ within 33 and 37°. When the theoretical analysis is repeated for the terminal state of the laminate the ultimate strength is estimated to be between 213 and 257 MPa with the upper bound close to the experimental value. Thirdly and finally, the ultimate strain measured experimentally is much higher than expected for polymeric composites, however consistent with the ply-closing mechanism (ref. 19).

## 5. CONCLUSIONS

* Experimentally, regular symmetrical laminates of the uniaxial, cross- and angle-plied constructions have been characterised for their stiffness and strength characteristics. The uniaxial construction has facilitated the establishment of lamina characteristics for the APC-2 system.

* Theoretically, the micromechanical and macromechanical concepts of the composite modelling have been utilised.

* Experimental findings have been correlated with the theory; The correlations have revealed:
  - The lamina characteristics (Engineering constants) can be predicted from the micromechanics with the accuracy better than 10%.
  - The laminate characteristics can be predicted from the macromechanics. Normalising on the predicted value, the experiment/theory correlation exhibits the following relative errors:

| LAMINATE | STIFFNESS | ULTIMATE STRENGTH |
|---|---|---|
| UNIAXIAL | -7.9 | not attempted |
| CROSS-PLIED | -0.3 | -11.0 |
| ANGLE-PLIED | +1.1 | + 8.6 |

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