DIMENSIONAL STABILITY EFFECTS IN THERMOPLASTIC COMPOSITES - TOWARDS A PREDICTIVE CAPABILITY

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ABSTRACT

Phenomena associated with thermal distortions in non-isotropic materials have been observed for many years, though little effort has apparently been made to derive a capability to accurately predict the magnitude of such distortions. The development of new high performance thermoplastic composites has led to increased awareness of these effects due to the high processing temperatures of such materials, and a number of workers have explained the effects in the simple case of slow, symmetrically-cooled laminates. This paper explores the possible contributing mechanisms to thermally-induced distortion, with particular reference to the effect commonly known as "spring-forward". It is shown that there are two separate cases which require analysis; slow, symmetric cooling and fast asymmetric cooling. In slow cooling of simple L-sections it is shown that most of the data required for the prediction of final part shape are available, and the magnitude of a number of second-order effects is explored. Fast cooling, and in particular fast asymmetric cooling typical of stamping operations, is discussed, and the development of a through-thickness gradient of residual stresses is examined. The ability of these stresses to produce "negative spring-forward" is demonstrated, and it is shown that experimental and analytical work can be combined to provide a predictive capability for simple shapes. Finally, the general direction of both current and future work is discussed.

INTRODUCTION

Continuous fibre reinforced composites offer the designer of structures the capability to provide strength and stiffness in areas where they are needed; their anisotropy in properties allows a structure to be designed efficiently, in a manner similar to that used by nature. However, material anisotropy can bring about problems during forming, when differences in coefficient of thermal expansion in and out of the plane may lead to distortions. This characteristic of anisotropic structural materials was examined independently by O'Neill [1] and Hamamoto [2], who showed that the thermal deformation of L-shaped components was a relatively simple function of initial part angle, coefficients of thermal expansion, and difference between forming temperature and observation temperature.

The introduction of PEEK based composites in the early 1980s [3], and the development of other, more recent tough thermoplastic materials [4] with relatively high processing temperatures has led to an increased awareness of the thermal distortion phenomenon. Early work in fabrication of such materials typically used processes which involved slow forming and cooling of components, and in general it was observed that the work of O'Neill could provide a good approximation of the overall distortion. However, more recent focus has been towards high rates of production for components, and the development of pultrusion and stamping technologies [5], where both forming and cooling rates can be high.

The work outlined in this paper shows that for rapidly formed Hercules AS4 continuous carbon fibre-reinforced PEEK (APC-2, supplied by ICI Thermoplastic Composites), induced distortions do not necessarily follow the simple form described by O'Neill. Indeed, several additional mechanisms contribute to the overall distortion of a fabricated component. These mechanisms are investigated and their effect evaluated, in particular, the mechanism relating to the residual stress distribution through the thickness of the laminate. Some preliminary work on the definition of these stresses is described, and a simple computational analysis of their effect is presented. It is shown that in simple cases, calculation of the processing induced distortions is possible, and that extension to more complex situations is in hand.

PROCESSING INDUCED DISTORTION - THE EFFECT OF MATERIAL ANISOTROPY

When processing anisotropic and orthotropic materials, it is commonly observed that the final shape of a part after removal from the mould is not necessarily identical to that of the mould itself [6]. Figure 1 illustrates the deformation which occurs. Polymer composites typically have a through thickness coefficient of thermal expansion which is much larger than that in the plane [7]. As a consequence, the large dimensional change which occurs in the thickness direction on cooling results in an apparent change of one radius of curvature of the bend with respect to the other. Low in-plane thermal expansion coefficients inhibit length changes in the arc sections, and the inevitable result is a reduction in enclosed angle as illustrated. This phenomenon is commonly referred to as "spring-forward".

O'Neill [2] showed that for any arbitrary curved section, the new angle could be calculated using the expression:

$$\Delta\theta_{th} = \theta(\alpha_z - \alpha_x)\Delta T \qquad (1)$$

where Θ is defined the sector angle, α denotes coefficients of thermal expansion, and ΔT temperature change.

In order to perform calculations based on O'Neill's analysis, the only experimental data required are accurate coefficients of thermal expansion, and a measure of ΔT. Coefficients of thermal expansion are easily determined, and are shown as a function of temperature for a laminate of the form $[45/-45/0/90]_{2s}$ in figure 2. Given the high value of through thickness coefficient of thermal expansion at high temperatures shown in figure 2 it is apparent that the accurate calculation of angular change using equation (1) will depend critically on the definition of the temperature at the onset of deformation. It is reasonable to suggest that the temperature at which the laminate is likely to begin deformation will be identical to that at which the material is first capable of sustaining residual stresses. This was explored using a technique described by Nairn and Zoller [8].

A series of unbalanced laminates of the form $[0_n/90_n]$ were heated slowly in a recirculating air oven, whilst the height of the centre of the laminate was monitored using a travelling telescope. The point at which the centre of the unbalanced laminate contacts the base of the apparatus (i.e. the temperature at which the radius of curvature becomes infinite) is the point at which the residual stresses have decayed to zero, and is defined as the stress-free temperature. A typical trace is shown in figure 3 in which the curvature is expressed in a non-dimensionalised form (laminate thickness divided by the curvature), and it is apparent that the stress free temperature for AS4/PEEK is approximately 310°C, which is in good agreement with the work of Nairn [8] and Parkyn [9].

Using experimental data for coefficients of thermal expansion shown in table 1 and figure 2, it is possible to perform a relatively simple incremental calculation of the total angular change expected in the case of a quasi-isotropic laminate manufactured from AS4/PEEK. For a simple part starting with an initial corner angle of 91.5°, a final part angle of 88.2° is calculated, reflecting a total angular change of almost 3.3°, as shown in table 2. (The use of an initial angle of 91.5° reflects analysis based on typical production tooling.) This is in excellent agreement with actual data for change in part angle, shown in table 3, for parts formed by a rapid diaphragm-forming process on a tool at 382°C, and cooled at an approximate rate of 20°C.min^{-1}. However, note that altering the stress-free temperature datum by +/-5°C can change the result of this calculation by approximately +1° or -0.3°, and so a degree of caution is necessary. Despite this, it can be seen that the calculation of deflections in this simple case is possible.

THE EFFECT OF RAPID COOLING

As well as the results of forming on a hot tool, angle changes measured for a series of L-sections formed on a room-temperature tool using the same rapid diaphragm forming technique are listed in table 3. In this case, the laminates have been subjected to rapid, asymmetric cooling. It is apparent that the spring-forward angle in this case is only 1.5°, which indicates that the situation can be rather more complex than that described by O'Neill and in the earlier work of Zahlan [6].

In a series of papers on residual stresses in thermoplastic composite materials, Chapman et al [10-12] showed that very rapid cooling of thick laminates can produce severe residual stress gradients. The form of the stress distributions was shown to be compressive on the external surface and tensile in the centre, and therefore unlikely to produce significant distortion. However, in the case of asymmetric rapid cooling there is no reason to consider that the stresses should be distributed symmetrically. The following section describes a method for examining these stresses.

THE DISTRIBUTION OF RESIDUAL STRESS THROUGH THE THICKNESS OF ASYMMETRICALLY COOLED LAMINATES

In order to simplify the analysis for the initial part of the work, the discussion in this section is restricted to laminates of the form $[0]_{16}$. Data covering the behaviour of multi-angle laminates, and a more complete description of the method will be reported at a later date [13].

Accurate estimation of in-plane residual stresses through the thickness of a composite laminate is most easily accomplished using the Process Simulated Laminate method described by Manson [14], and more recently Chapman [15]. Using this technique layers of polymeric release film are incorporated in the body of the laminate, and after manufacture the stack is separated. The change in curvature of the laminate, or the change in strain at the surface as a function of thickness can be used to calculate the stress release associated with layer removal.

Flat Laminates

In this series of experiments a number of 300mm square laminates of the form $[0]_{16}$ were prepared using AS4/PEEK, with a 50μm Upilex S polyimide film separating each group of four plies. Thus each complete laminate was made up of four sublaminates. Using the same rapid pressure-forming machine utilised for manufacture of the L-sections described above, the laminates were consolidated onto a flat, steel tool, which was maintained at fixed temperatures, shown in table 4, during the start of the forming cycle. Temperature was monitored at six positions through the thickness of each laminate; for the most severe case, that for a panel moulded onto a 37.8°C (100°F) tool, cooling rates for the laminate as a whole were typically $650°C.min^{-1}$ over the critical region from the melt to the Tg (143°C). Most important is that a temperature differential of between 50°C and 70°C was maintained through the thickness of the laminate between solidification and the Tg, leading to the generation of residual stresses as described by Chapman [10]. Comparison of the data with those from laminates containing no polymeric interlayers confirms that the effect of the Upilex was measurable but small, as also observed by [10]. Laminates manufactured on tools held at 37.8°C (100°F) and 149°C (300°F) were found to exhibit significant curvature in the direction transverse to the fibres after removal, whilst those manufactured on a 260°C (500°F) tool were found to possess zero or slight negative curvature. The measured radii of curvature for typical laminates are also shown in table 4.

After manufacture, the laminates were cut into strips approximately 10mm wide, and strain gauges were bonded to the surfaces with the laminates held in the flat condition. The change in local strain at the gauges was then recorded as successive layers were removed. It is interesting to note that each of the sublaminates from the panels produced on the two low temperatures tools retained significant curvature after disassembly.

Using an analysis reported by Chapman [15] based on the earlier work of Lee and Rogers [16] and Treuting and Reid [17], the strain gauge data can be reduced to equivalent stresses using the equation:

$$\sigma_2 = -\frac{E_2}{2}\left(\left(\frac{d}{2}-z\right)\frac{(\Delta\epsilon_s)}{(\Delta z)} - 4\epsilon_s(z) + 6\left(\frac{d}{2}-z\right)\sum\frac{(\epsilon_s\phi)}{\left(\frac{d}{2}-\phi\right)^2}\Delta\phi\right) \quad (2)$$

Where z is the distance of the removed layer from the centre of the laminate, d is laminate thickness, ϵ_s is the absolute value of strain at the surface, and $\Delta\epsilon_s$ is the change in surface strain with the removal of a layer thickness Δz. In the summation term, the terms in ϕ are equivalent to $\Delta\epsilon_s$, Δz and z for each sublaminate removed. It was found that for several laminates formed under identical conditions, the differential value of stress through the thickness remained constant, though the absolute values at the surfaces could vary significantly. The form of the data presented here are in agreement with those of other workers [18], though a more thorough exploration of this effect is essential.

Hence the residual stress was calculated as a function of position in the laminate: For brevity, only the most severe case will be presented here.

In figure 4 the distribution of residual stress through the laminate manufactured on a 37.8°C (100°F) tool is shown. It is important to note that since the initial starting point for the strain was a laminate held flat, the stresses represented in figure 4 are those which cause the curvature to occur on cooling, not those which exist in the cooled laminate in the unrestrained condition. It is interesting that the stresses shown in figure 4 were calculated from a starting point on the surface furthest from the tool, rapidly-cooled, surface, and therefore the stresses are forced to be tensile, since in the flat condition the upper surface is under tensile strain. If the opposite face is used as a starting point, the stresses are reversed in magnitude and direction, but result overall in identical deflection of the plate.

In order to test that the magnitude of the stresses shown in figure 4 is indeed realistic, a simple two-dimensional finite element (FE) model of a beam was created, using 4-noded elements, with four elements through the thickness of the laminate. Strains calculated from the stresses shown in figure 4 were applied to the model in two ways: in the form of tractions along the interfaces between each of the sublaminates and on each surface, and also as point deflections on the ends of the beam. In each case the resultant deflection of the beam was found to match that of the real case within 10%.

L-Section Laminates

The stresses measured in the laminate were shown to be capable of causing the deflections observed. The same tractions and point deflections were then applied numerically to a finite element model of an L-section, constructed using the same elements as those used in the beam model. Figure 5 shows the undeformed and deformed L-section model, with the fibres oriented along the edge of the corner, in the z-direction. It is apparent that the numerical model predicts that an L-section of this type should exhibit an angle increase, although the magnitude of this increase was found to vary depending upon the loading condition. In the case of imposed tractions, the angular increase calculated was 0.6° for an initial corner angle of 91.5°, whilst the end-deflection condition was found to result in a greater increase of approximately 2°. Whilst each of the techniques for application of dimensional changes has it's respective merits, it is believed that the former case is more appropriate.

Although the model described above does not reflect a realistic composite component lay-up, a simple L-section with fibres running parallel to the corner is the most severe condition for residual-stress-gradient induced deflections. In order to test that such "negative spring-forward" is possible, an L-section of the type described in the model, and containing Upilex layers, was moulded on a 37.8°C (100°F) tool with a 91.5° corner angle. Figure 6 shows a section through the laminate, and an angle increase of approximately 2.2° was recorded. In this case, the distortion due to differential coefficients of thermal expansion should be minimal. Previous work has shown that the through-thickness coefficient of thermal expansion of AS4/PEEK is greater than that in the plane [9], but in the uniaxial case the discrepancy is of the order of 5%. Hence an incremental calculation of the type shown in table 2 predicts an angular decrease of 0.015°.

This is in relatively poor agreement with the preferred FE result, though at the present time it is encouraging that the phenomenon of angular increase is both predicted and observed in this rather idealised case. It is apparent that further refinement is required, since it has not been possible to identify an alternative mechanism for the production of an angular increases in the process of fabrication.

THE EFFECT OF PLY GEOMETRY

The flow processes which take place during the fabrication of thermoplastic based composites can lead to significant changes to the geometry of the laminate plies. In particular, individual plies can change in thickness due to polymer flow under pressure. Such geometry changes influence the thermal distortion through two mechanisms: changes to the thermal expansion characteristics of the material, thereby altering the anisotropy of properties applied in equation (1); and changes to the force equilibrium within the laminate, resulting in stress-induced out-of-plane curvature. The contribution of these distortion mechanisms to the overall dimensional stability can be predicted using the finite element method.

Typically, thinning in not a uniform phenomenon in that only fibres running parallel to the corner are likely to flow, and thinning is usually most severe on the outer plies of the corner. Two L-section components formed at fast and slow forming rates have been sectioned and the resulting ply geometry determined. The geometry of the two sections, and one of uniform ply thicknesses, have been modelled, using coarse finite element representations, and the distortion of the enclosed angle predicted. The model geometries illustrating the thinning are shown in figure 7. Thermo-elastic properties applied within the models were calculated using classical laminate theory for in-plane properties and volume-weighted averaging for those out-of-plane [19]. For the purpose of this preliminary modelling, the input properties were assumed to be constant throughout temperature range. Therefore, it is the relative, rather than absolute, values of the results which are significant.

Two types of models have been generated for each case. In the first, uniform, homogeneous, effective properties were assumed for the material of the L-section; thereby, isolating the influence of the thermal property anisotropy and eliminating that of non-symmetry of the plies. In the second approach, each ply was represented individually; so that both the thermal property anisotropy and the non-symmetry effects were superposed.

The predicted changes in thermal properties and L-section angles are listed in table 5. The influence of the non-symmetry of plies is represented by the difference in angle change predicted by the two models. Although both distortion mechanisms are influenced by the change in ply geometry, it is clear that the effect of non-symmetry of the laminate is more significant than the change in through-thickness coefficient of thermal expansion. It is interesting that the ply thinning which occurred in the fast formed laminate results in a reduced change in total angle, whereas that occurring in the slow formed laminate increases this distortion. Clearly, the effect of ply geometry contributes to thermal distortion both positively and negatively depending on the individual case.

Although further investigation is required to enable accurate prediction of the influence of ply geometry of the dimensional stability of fabricated components, the ability to estimate these effects has been demonstrated and the mechanisms can be ranked in importance with others discussed elsewhere in this paper.

DISCUSSION AND ADDITIONAL EFFECTS

The work above suggests that the total fabrication induced angle change in a thermoplastic composite is the cumulative result of several distortion mechanisms. If calculation of spring-forward by summation is permissible, then for the case of forming a simple two-dimensional component, the total angular distortion may be given by:

$$\Delta\theta_{total} = \Delta\theta_{thermal} + \Delta\theta_{stress} + \Delta\theta_{imbalance} \tag{3}$$

Here, the first two terms may have roughly equivalent value, whilst the last term is likely to be significantly smaller.

While this expression provides a key to the distortion of a section at the end of the initial fabrication cycle, it is recognised that thermoplastic composites such as AS4/PEEK are often subjected to some post-forming heat treatment during bonding operations. Most common is the use of amorphous interlayer bonding [20], in which the composite is heated to approximately 300°C (572°F). In this case there is the potential for additional distortion of the part due to two effects - changes in crystallinity and stress relaxation.

Composites based on PEEK have been shown to be tolerant to a very wide variation in processing cycle without changing final performance [21,22]. Following rapid cooling, crystallinity gradients can be produced in the material [10]. On subsequent heating above approximately 200°C (392°F), further crystallisation is likely, with resulting local volume reduction [23]. A simple analysis based on the consideration of these volume changes as an analogue of coefficient of thermal expansion reveals that for a uniform change in crystallinity of 1% through the thickness of a quasi-isotropic laminate, the resulting change in angle of a 90° bend is approximately 0.02°. A full description of this work will be published at a later date [24], but it's inclusion here illustrates the magnitude of the effect.

While the effects of constraint during post-process heating have received little attention in previous work, this is now considered to be one of the major contributors to difficulties in part assembly. Though few hard data are available, Humbertson [25] has observed that in simple L-sections formed onto melt-temperature tools (thus minimising through-thickness stress effects), restraining the corner angle during heating to 315°C (600°F) can result in an additional spring-forward of up to 1.1°. Similar restraint of a part formed on a 37.8°C (100°F) tool during heating to the same temperature can cause up to 2° of additional distortion, though the final angle of the component was found to be close to that of the previous case. The implication here is that two mechanisms are possible - relaxation of any residual stresses gradient through the thickness (as also observed by Chapman [10]) and an analogue of creep forming under the restraining effects of locating tools. Hence the separate effects of post processing can be conveniently summarised thus:

$$\Delta\theta_{additional} = \Delta\theta_{relaxation} + \Delta\theta_{creep} + \Delta\theta_{recrystallisation} \tag{4}$$

While expressions (3) and (4) are not rigourous, we belive that they provide the conceptual basis for predicting the complete distortion of a simple component due to fabrication effects. The ranking of the magnitude of each effect appears highly reasonable, and each of the components described is currently under investigation. At the present time, the final shape of a simple L-section, or other component with two dimensional curvature appears to be predictable after forming, though post-forming effects are more difficult to assess. The extension of these analyses to full three dimensional shaping is non-trivial, and will require considerable effort to complete; however, it is believed that the work presented here outlines the foundation upon which such calculation can be built.

CONCLUSIONS

We have identified six mechanisms which contribute to the phenomenon of spring-forward in thermoplastic composites laminates. Three of these occur during the process of cooling from melt, whilst those remaining may be the result of post-forming operations. The six mechanisms are:

- Anisotropy of thermal properties, in and out of plane.
- In-plane residual stress gradients through the thickness.
- Ply geometry changes resulting in force imbalance.
- Internal stress relaxation during post forming heating.
- Creep of constrained components during post forming operations.
- Recrystallisation during post forming operations.

The most significant of the above mechanisms are those relating to anisotropy in thermal expansion properties, and the creation of in-plane stresses varying through the thickness due to asymmetric rapid cooling in certain processing operations. The magnitude of each of these effects is estimated in this paper.

The effects of imbalance due to local thinning in processing and further crystallisation during heating are discussed and are shown to be second order effects. The processes of stress relaxation and creep in post forming operations are briefly discussed and available data suggest that these effects may be highly significant.

The intention of this paper has been to identify these components and define their magnitude. Further work is required to fully define the interaction between the contributing phenomena.

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Temp. (°C)	α_z x10^{-6}	α_x x10^{-6}
310	372.83	8.17
290	270.89	8.17
270	208.43	8.17
250	172.23	5.93
230	150.99	5.93
210	135.06	5.93
190	124.50	4.67
170	115.86	4.67
150	95.50	3.41
130	68.49	3.41
110	50.36	3.41
90	47.17	3.41
70	46.98	3.41
50	45.81	3.20
30	43.55	3.20

Table 1: Change in coefficient of thermal expansion with temperature in-plane (α_x) and through thickness (α_z) for a laminate of the form $[+45/-45/0/90]_{2S}$.

Temp. (°C)	α_z x10^{-6}	α_x x10^{-6}	Incremental Springfwd. (Degrees)	Total Springfwd. (Degrees)	Corner Angle (Degrees)
310	372.83	8.17	0.000	0.000	91.500
290	270.89	8.17	0.645	0.645	90.854
270	208.43	8.17	0.468	1.114	90.386
250	172.23	5.93	0.359	1.473	90.027
230	150.99	5.93	0.299	1.772	89.728
210	135.06	5.93	0.262	2.034	89.466
190	124.50	4.67	0.234	2.268	89.232
170	115.86	4.67	0.216	2.485	89.015
150	95.50	3.41	0.202	2.688	88.812
130	68.49	3.41	0.168	2.856	88.644
110	50.36	3.41	0.119	2.974	88.526
90	47.17	3.41	0.086	3.060	88.440
70	46.98	3.41	0.080	3.140	88.359
50	45.81	3.20	0.079	3.220	88.280
30	43.55	3.20	0.078	3.299	88.201

Table 2: Change in α and Θ with temperature.

Part	Tool Temp. °C (°F)	Part Angle (Degrees)	Angle Change (Degrees)
A1	382 (720)	88.33	3.17
A2	382 (720)	88.33	3.17
A3	382 (720)	88.67	2.83
A4	382 (720)	88.50	3.00
A5	382 (720)	88.25	3.25
	mean	*88.42 (0.17)*	*3.08 (0.17)*
B1	382 (720)	88.25	3.25
B2	382 (720)	88.00	3.50
B3	382 (720)	88.00	3.50
B4	382 (720)	88.17	3.33
B5	382 (720)	88.17	3.33
	mean	*88.12 (0.11)*	*3.38 (0.11)*
D1	24 (75)	90.00	1.50
D2	24 (75)	90.00	1.50
D3	24 (75)	90.00	1.50
D4	24 (75)	90.00	1.50
D5	24 (75)	90.00	1.50
	mean	*90.00 (0.0)*	*1.50 (0.0)*

Table 3: The effect of tool temperature (and hence cooling rate) on spring-forward. Standard deviations in parentheses.

Temperature (°C)	Radius of curvature (m)
37.8	0.62
149	0.94
260	Flat

Table 4: The effect of tool temperature on plate curvature.

GEOMETRY		UNIFORM	FAST	SLOW
α_x	x10^{-6} /°C	2.70	2.54	2.29
α_z	x10^{-6} /°C	40.93	40.92	40.90
ΔT	°C	300	300	300
$\Delta\theta - FE - Homogeneous$	DEG	1.036	1.040	1.044
$\Delta\theta - FE - Layered$	DEG	1.050	1.014	1.119

Table 5: The effect of ply geometry on thermal properties and on change in angle, predicted by FE analysis.

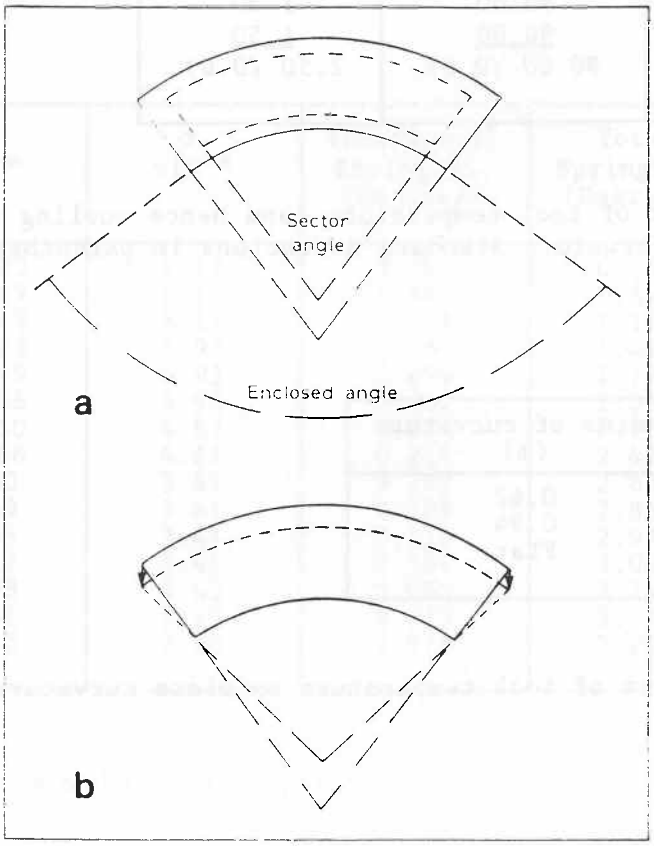

Figure 1: Thermally induced distortion in an arc section of a) isotropic material, and b) anisotropic material.

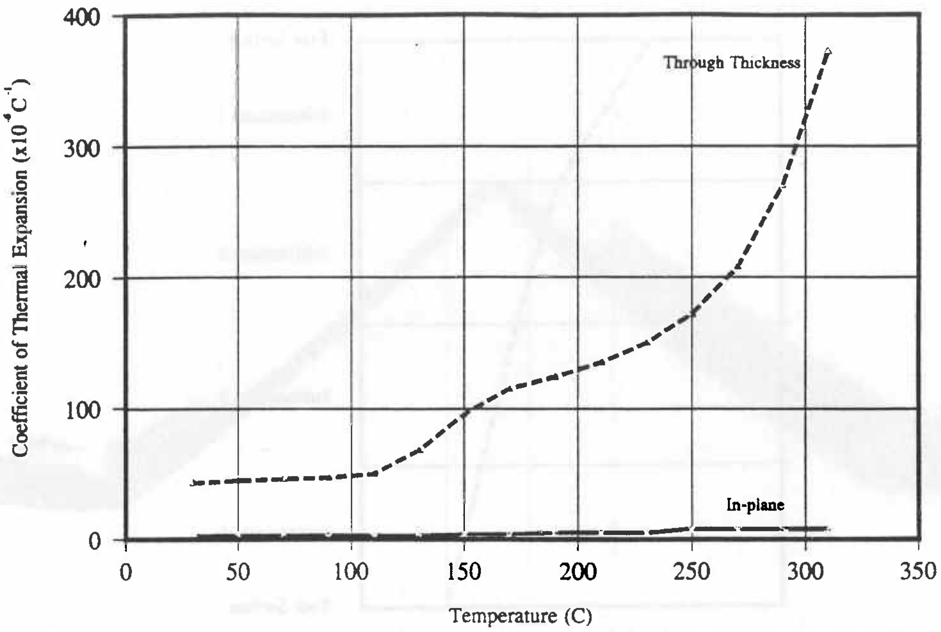

Figure 2: Change in coefficient of thermal expansion of a quasi-isotropic AS4/PEEK plate with temperature.

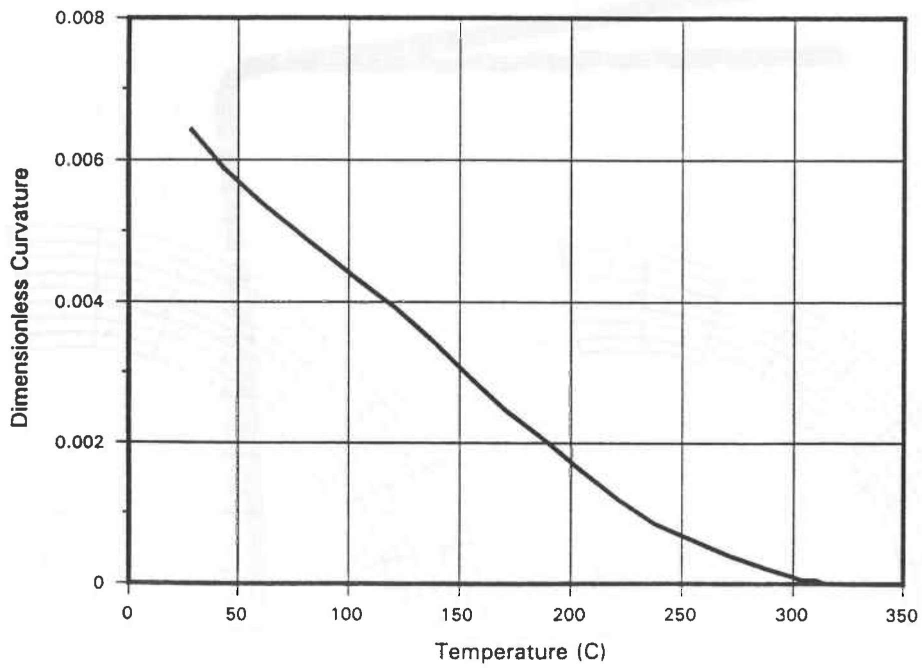

Figure 3: Change in curvature of a [0/90] PEEK composite laminate with temperature.

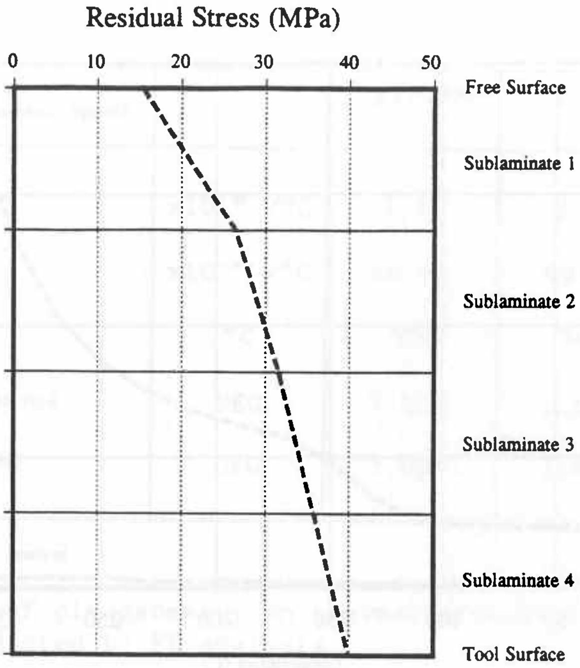

Figure 4: Stress distribution through sublaminates determined by the process simulated laminate method.

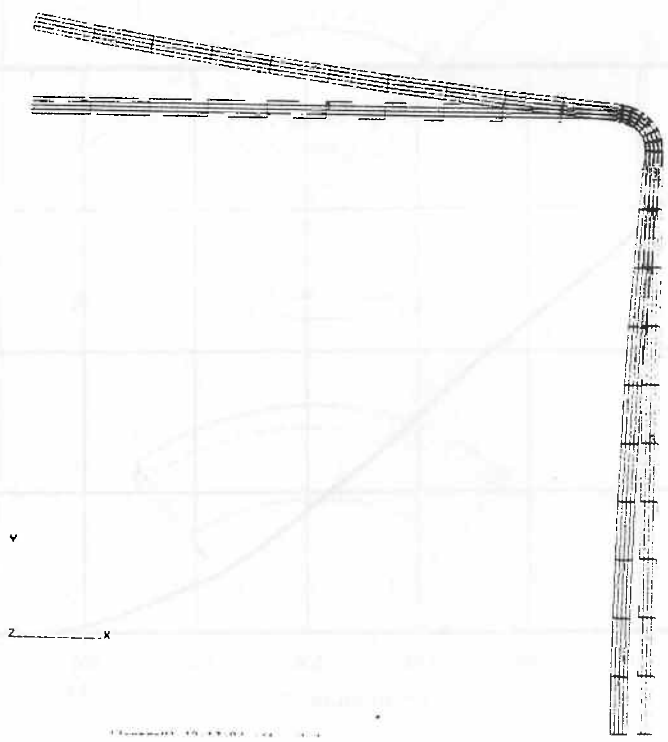

Figure 5: Finite element model of loaded and unloaded L-section.

Figure 6: Cross-section through L-section moulded on a room-temperature tool.

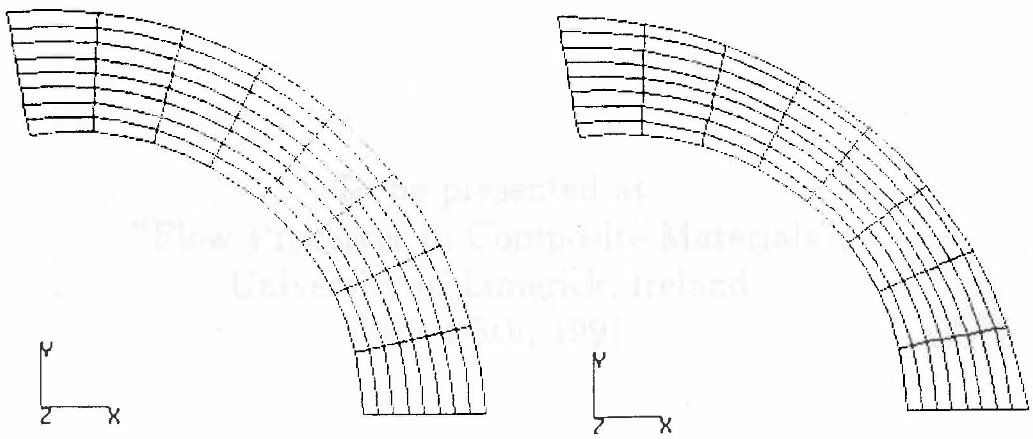

Figure 7: Deformed model geometries of fast formed and slow formed L-sections illustrating the ply thinning.