THE EFFECT OF PRESSURE GRADIENTS ON THE CONSOLIDATION OF FIBRE-REINFORCED THERMOPLASTICS

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ABSTRACT

The effect of the applied pressure, preform thickness and mould geometry on pressure gradients during the forming of unidirectional PEEK/carbon fibre and woven PEI/carbon fibre step-tapered laminated plates was investigated. The evolution of pressure and temperature during processing was monitored and microscopy was used to determine the degree of consolidation of the laminates. The principle factors influencing the magnitude of the pressure gradients were fibre orientation for PEEK/CF and applied pressure for PEI/CF (weave). The influence of the pressure gradients on consolidation quality was determined and a model of the consolidation process was proposed for unidirectional and woven fibre-reinforced thermoplastics. This model can be used to estimate part quality.

1. INTRODUCTION

Continuous fibre-reinforced thermoplastic composites have become an increasingly important class of materials over the last decade, gaining significant acceptance in high-performance applications. They are advanced materials that provide good performance and controlled properties through high fibre volume fractions and well-defined fibre alignment. In many cases the processing technology of advanced thermoplastic composites has mirrored that of advanced thermosetting composites, which have been in widespread industrial use for over twenty years. Thermoplastic-based composites offer several unique processing capabilities, in that they may be formed and/or reprocessed by the reapplication of heat and pressure. Complex-shaped three-dimensional parts may be formed in several stages; parts can be thermally joined to produce monolithic composite assemblies; reconsolidation can be used to eliminate imperfections such as cracks, voids and dimensional

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inaccuracy. Moreover thermoplastic-based parts have potential for recycling. These unique features lead to new processing techniques which in turn stimulate industrial use of thermoplastic-based composites, as cost effectiveness can be ensured through simplified and rapid net-shape production with a minimal trade-off in performance.

The properties of polymer-based composites depend strongly on the microstructure induced in the laminate during processing. For instance, it has been observed that voids resulting from the application of insufficient pressure during processing alter the composite properties and the final part quality [1,2]. It is therefore of prime importance to obtain parts having a final microstructure of good quality. Several processing phenomena affect the part quality: deconsolidation and reconsolidation behaviour, matrix degradation, and pressure gradients. The influence of these factors on the consolidation behaviour and the consolidation mechanisms responsible for these voids must be analysed if optimal processing conditions are to be defined.

The interest in manufacturing composite parts having a complex geometry is steadily increasing. Press forming is a promising technique to achieve this goal. However, it may lead to pressure gradients within the part during consolidation, as shown in Figure 1. The part is subjected to a range of pressures during moulding depending on its geometrical configuration. Thus, some locations in the part may be submitted to a pressure lower than the applied pressure inducing only a partial consolidation. Consequently, it is of prime importance to define the parameters affecting these pressure gradients.

This study focused on the effect of pressure gradients on the quality of press-formed step-tapered laminated plates. The influence of preform thickness, fibre structure and orientation, step height, and applied pressure on pressure gradients and part quality were investigated.

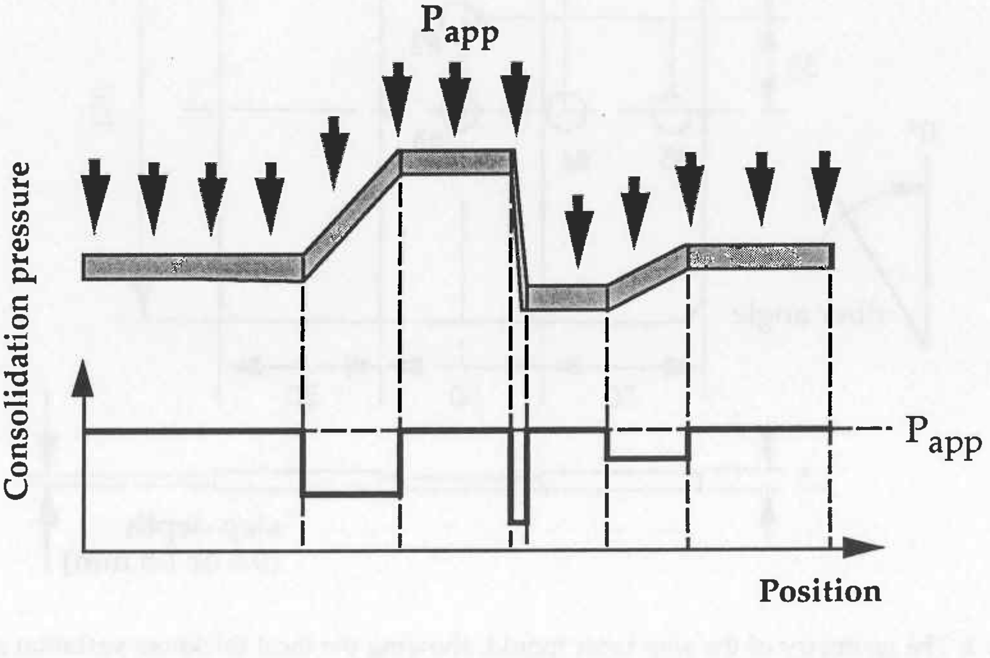

Figure 1: Schematic pressure distribution within a complex three-dimensional part

2. EXPERIMENTAL

The materials used in this study were unidirectional carbon fibre-reinforced PEEK (PEEK/CF, APC2/AS4 from ICI-Fiberite) and woven carbon fibre-reinforced PEI (PEI/CF (weave), CETEX from Ten Cate). These materials have fibre volume fractions of 61% and 58%, respectively.

Laminated preforms were processed in a 150 x 150 mm² matched-die mould mounted on a Schwabenthan Polystat 400S instrumented press. Resin flow was negligible. The thickness of the part in the central zone of the mould was increased by varying the vertical position of the central section of the lower mould half, as shown in Figure 2. The pressure gradients resulting from this step taper represent those obtained in parts of constant thickness but of varying angle. Five contact pressure transducers for direct pressure and temperature measurement were set in the upper mould, flush to the mould surface at locations #1 - #5 (Figure 2).

The prepreg plies were heated to their processing temperature in the mould, under contact pressure at a heating rate of 2.4°C/min. At the processing temperature, the processing pressure was applied and maintained until the end of cooling to avoid deconsolidation. During processing, pressure and temperature were recorded using an HBM-DMC data acquisition system. The processing cycle is illustrated in Figure 3.

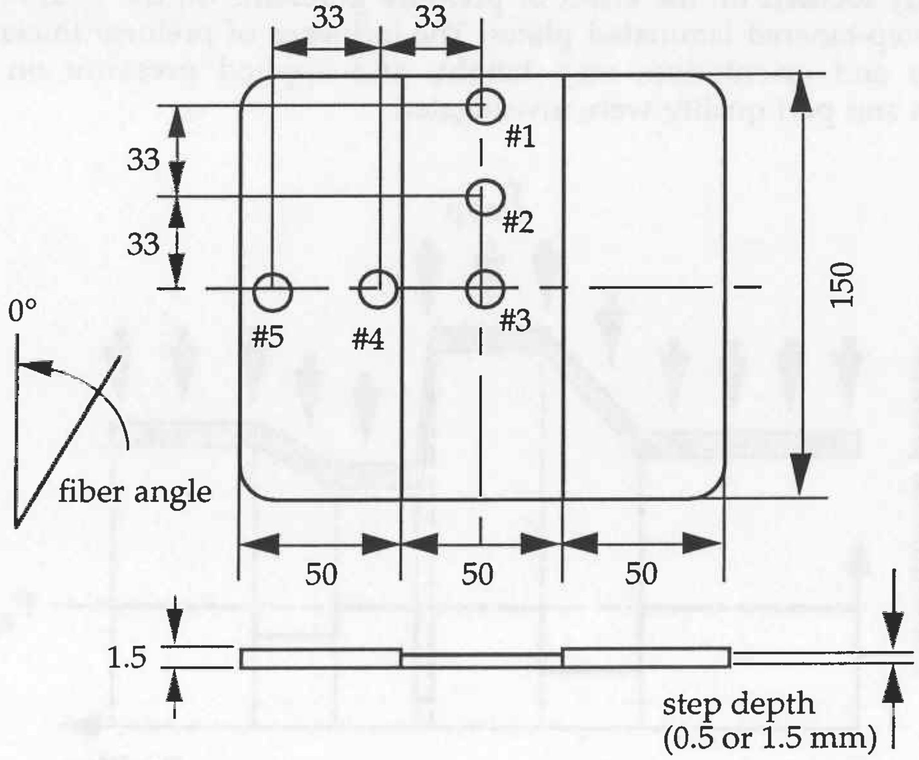

Figure 2: The geometry of the step taper mould, showing the local thickness variation and fibre orientation relative to the central section.

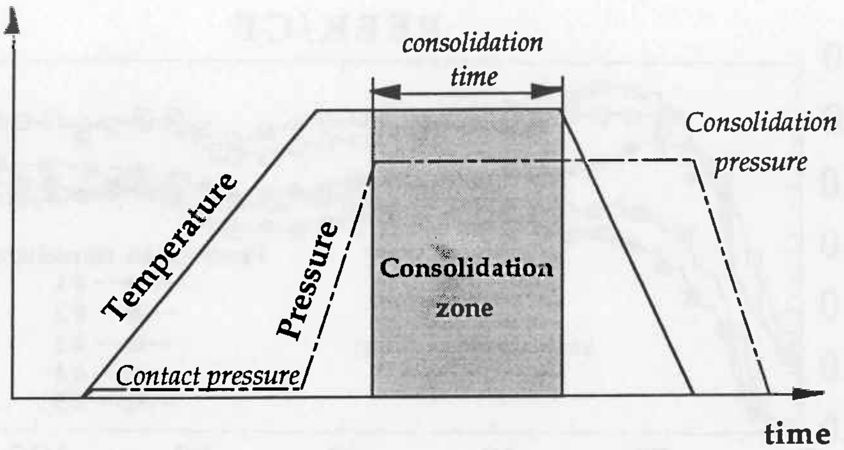

Figure 3: Schematic representation of the processing cycle.

The pressures at the five locations were measured as a function of the preform thickness, fibre orientation, step depth and applied pressure. Table 1 lists the test conditions for the two materials.

Table 1: Process parameters for the determination of pressure gradients.

Parameters	**PEEK/CF**	**PEI/CF (weave)**
Number of plies	16, 24	8, 12
Fibre orientation	0°, 45°, 90°	0°/90° (fabric), ±45°
Central section depth (mm)	0.5, 1.5	0.5, 1.5
Processing temperature (°C)	380	360
Pressure P_{app} (bar)	10, 40	10, 40
Consolidation time (s)	120	120

To observe the effect of pressure gradients on part quality, related to the development of intimate contact and fibre impregnation, microstructural analysis was performed on the samples after demoulding using an optical microscope.

3. RESULTS AND DISCUSSION

3.1 Influence of composite and mould design on pressure gradients

Figure 4 shows the evolution of the pressure at each transducer location during the consolidation of 16 layers of PEEK/CF with a fibre orientation of 0° and a step depth of 1.5 mm at 10 bars.

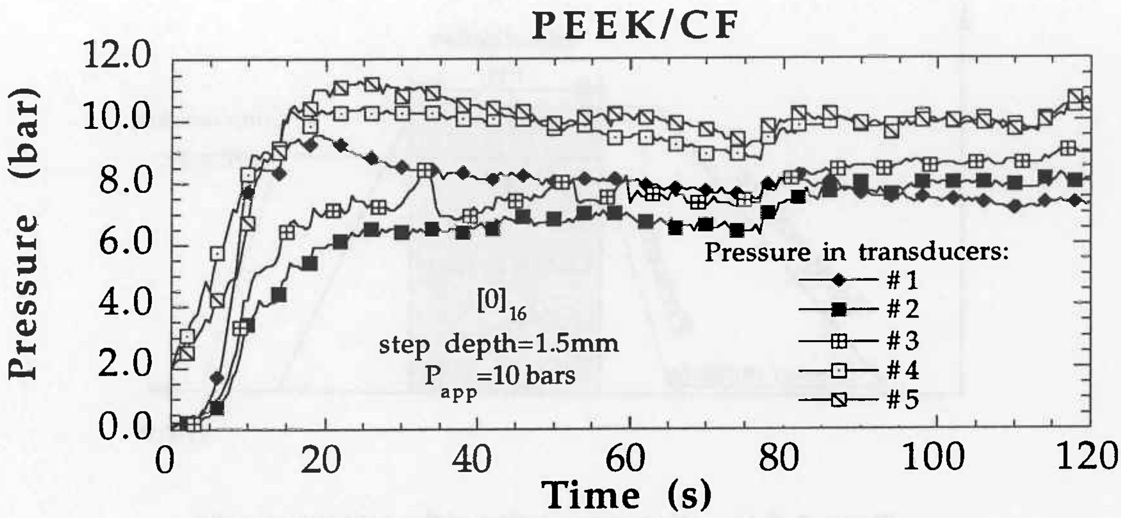

Figure 4: Evolution of pressure at positions #1-5 during consolidation of 16 layers of PEEK/CF with a fibre orientation of 0° and a step depth of 1.5 mm at 10 bars.

The pressure efficiency (PE) inside the sample, defined as the ratio of the pressures measured at transducer locations #3 and #4, was determined from experimental data such as that shown in Figure 4. Thus a homogeneous pressure distribution (no pressure gradients) throughout a part means a high PE, and a more efficient processing cycle (Figure 5).

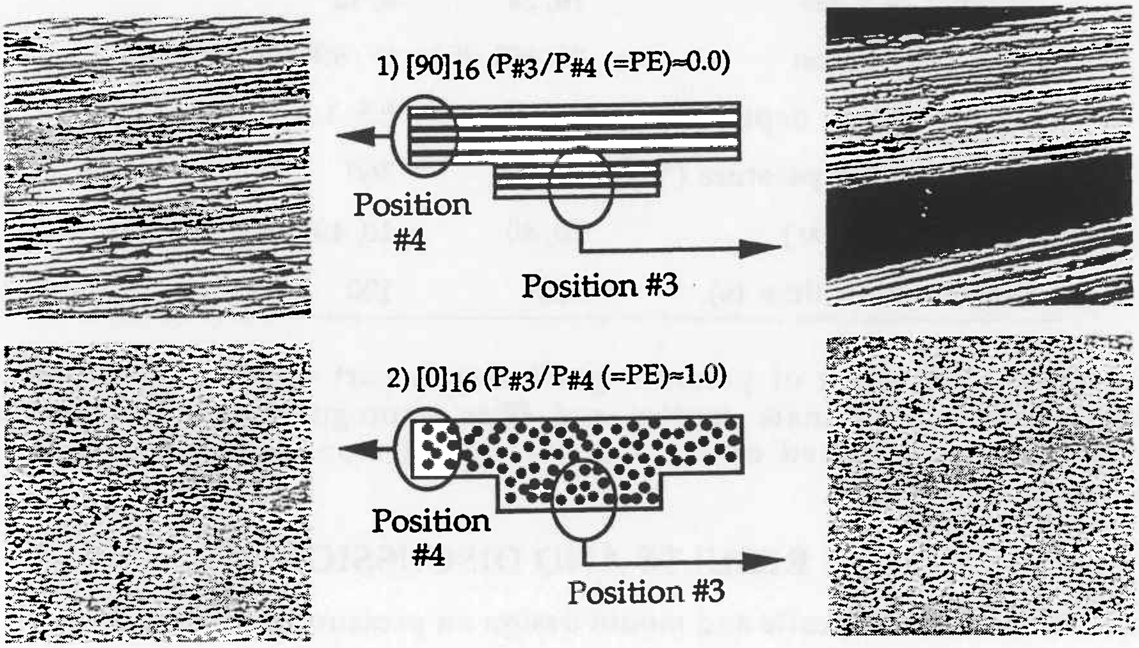

Figure 5: Microstructure and pressure efficiency (PE) of 1) $[90]_{16}$ and 2) $[0]_{16}$ PEEK/CF processed at 380°C under 10 bars with a consolidation time of 120s and a step depth of 1.5mm.

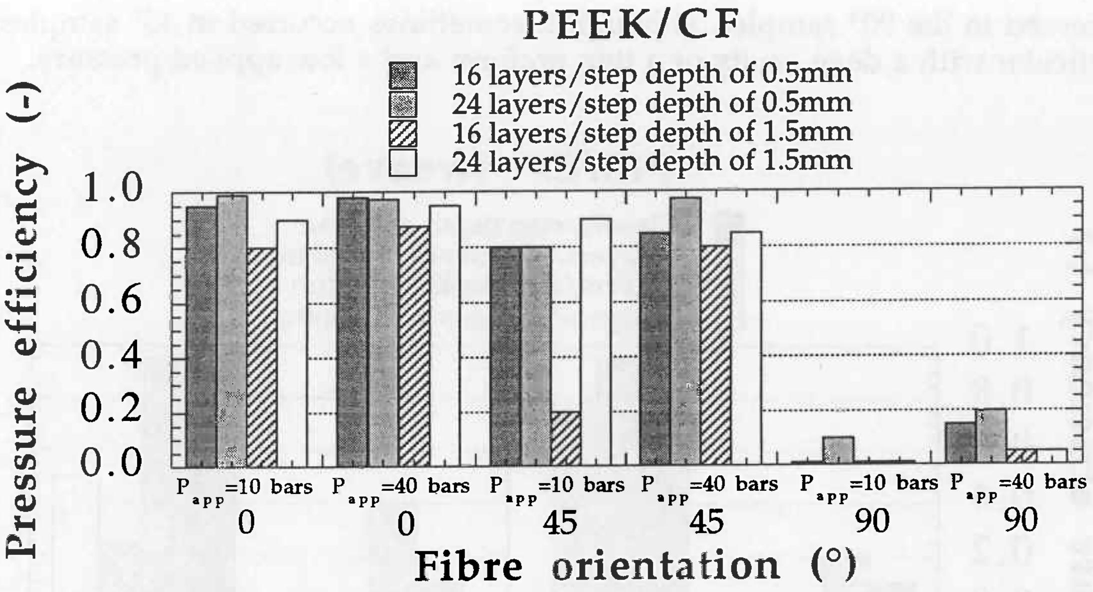

Figure 6: Pressure efficiency as a function of the processing parameters for PEEK/CF.

The variation of the pressure efficiency versus preform thickness, fibre orientation, step depth and applied pressure is shown in Figure 6 for PEEK/CF and in Figure 7 for PEI/CF (weave).

From Figure 6 it can be concluded that for PEEK/CF:

- a high PE (>0.8) was achieved for a fibre orientation of 0° and 45°,
- neither the preform thickness nor the step depth had a strong influence on the magnitude of the PE,
- a low PE (<0.2) was achieved for a fibre orientation of 90° since inextensible fibres cannot easily fill the step zone of the mould.

From Figure 7, it can be concluded that for PEI/CF (weave):

- a low PE (<0.2) was achieved for an applied pressure of 10 bars, and neither the preform thickness nor the fibre orientation exerted a strong influence on the magnitude of the PE,
- a high PE (>0.7) was achieved for an applied pressure of 40 bars independently of the fibre orientation and preform thickness,
- as the step depth increased, the PE decreased drastically and approached zero, except for a fibre orientation of 0/90° (fabric) and an applied pressure of 40 bars.

The effect of fibre orientation was more pronounced for PEEK/CF than for PEI/CF (weave). Furthermore, fibre buckling was observed for PEI/CF (weave) samples with a fibre orientation of 0°/90°, but this phenomenon disappeared for a fibre orientation of ±45°. With PEEK/CF, fibre buckling was mainly observed in the 90° samples, although it sometimes occurred in 45° samples, in particular with a deep cavity or a thin preform and a low applied pressure.

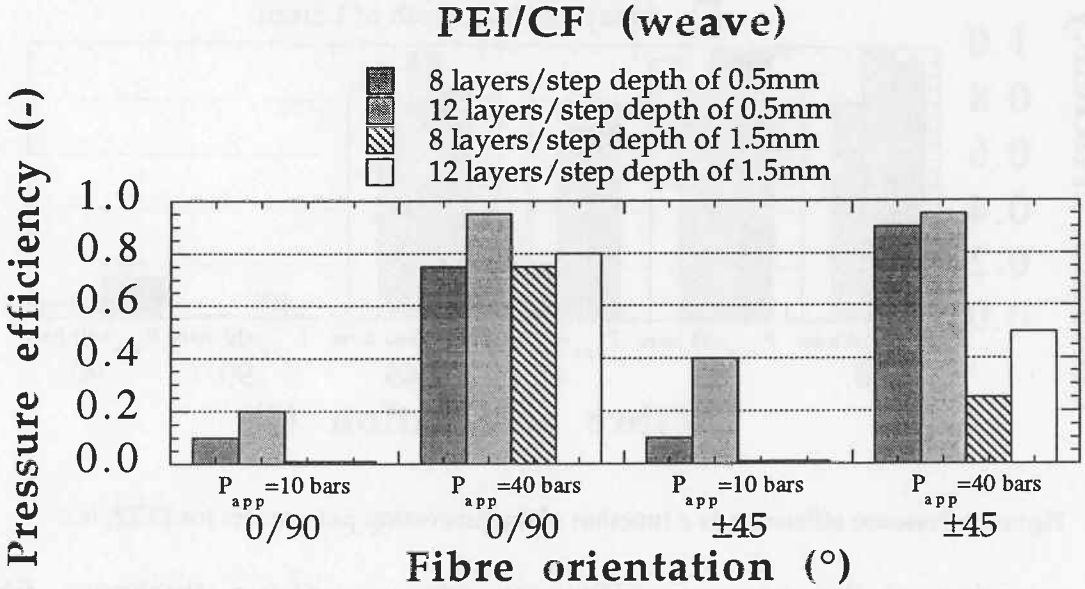

Figure 7: Pressure efficiency as a function of the processing parameters for PEI/CF (weave).

3.2 Prediction of the consolidation behaviour versus pressure gradients

The consolidation behaviour of a step-tapered laminated plate can be predicted using a consolidation model together with the experimental pressure measurements. The consolidation of thermoplastic matrix composites consists of three major steps:

- intimate contact,
- autohesion
- fibre impregnation.

These mechanisms are schematically represented in Figure 8. In this study, autohesion was not considered. It was shown in a previous study that autohesion is almost instantaneous for both materials at the temperatures commonly used for processing [3].

Since the surfaces of successive plies are rough, gaps exist between the plies prior to the application of heat and pressure and the matrix must be deformed to produce intimate contact between the plies. Lee *et al.* have represented the irregular ply surface by a series of equisized rectangles [4,5]. As a result of the applied pressure the rectangular elements spread along the interface, as shown schematically in Figure 9.

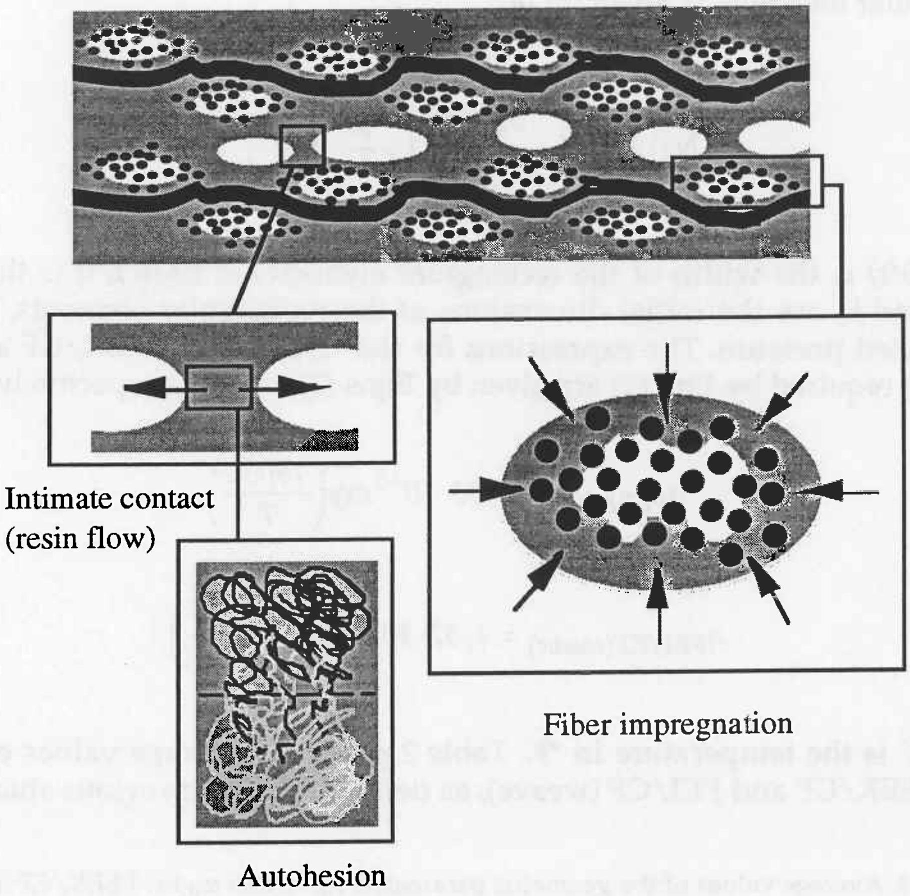

Figure 8: Schematic of the mechanisms involved in consolidation: intimate contact, autohesion, fibre impregnation.

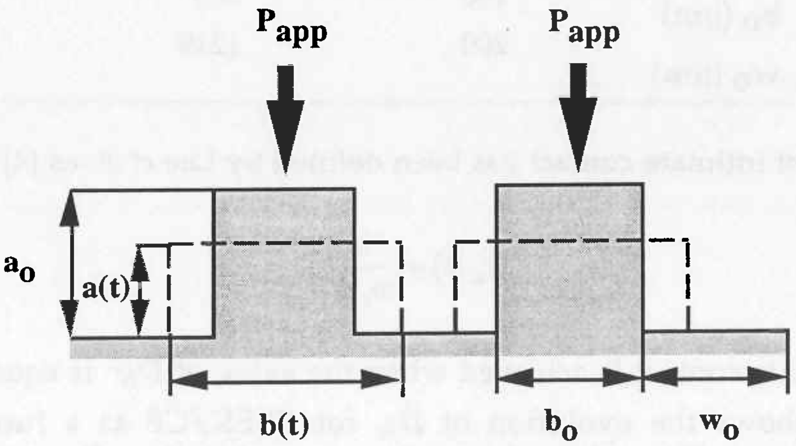

Figure 9: Spreading of rectangular elements as a result of applied pressure.

Based on this geometry, the evolution of the viscous deformation of the rectangular elements is given by [4]:

$$b(t) = b_0 \left[1 + \frac{5 P_{app} t}{\eta} \left(1 + \frac{w_0}{b_0} \right) \left(\frac{a_0}{b_0} \right)^2 \right]^{\frac{1}{5}} \tag{1}$$

where $b(t)$ is the width of the rectangular elements at time t, η is the viscosity, w_0, a_0 and b_0 are the initial dimensions of the rectangular elements, and P_{app} is the applied pressure. The expressions for the viscosity of PEEK/CF and PEI/CF (woven) required by Eqn (1) are given by Eqns (2) and (3), respectively [3,6]:

$$\eta_{PEEK/CF} = 5{,}66 \cdot 10^{-8} exp\left(\frac{19032}{T} \right) \tag{2}$$

$$\eta_{PEI/CF(weave)} = 4{,}52 \cdot 10^{-11} exp\left(\frac{20275}{T} \right) \tag{3}$$

where T is the temperature in °K. Table 2 gives the average values of a_0, b_0 and w_0 of PEEK/CF and PEI/CF (weave), as determined in a previous study [3,7].

Table 2: Average values of the geometric parameters a_0, b_0 and w_0 for PEEK/CF and PEI/CF (weave).

Parameter	PEEK/CF	PEI/CF (weave)
a_0 (µm)	50	224
b_0 (µm)	150	603
w_0 (µm)	200	1219

The degree of intimate contact has been defined by Lee *et al.* as [4]:

$$D_{ic}(t) = \frac{b(t)}{w_0 + b_0} \tag{4}$$

where intimate contact is achieved when the value of D_{ic} is equal to unity.

Figure 10 shows the evolution of D_{ic} for PEEK/CF as a function of time, material location, fibre orientation and pressure, based on Eqns (1) and (4). At a fibre orientation of 90°, the pressure distribution within the part was not homogeneous (PE<0.2) and the part was only partially consolidated. The pressure at location #3 was too low to consolidate the material in this region, even for an applied pressure of 40 bars. However, for a fibre orientation of 0° the pressure distribution within the part was close to homogeneous (PE>0.8), and the part was well consolidated. The predictions correlate well with the micrographs presented in Figure 5.

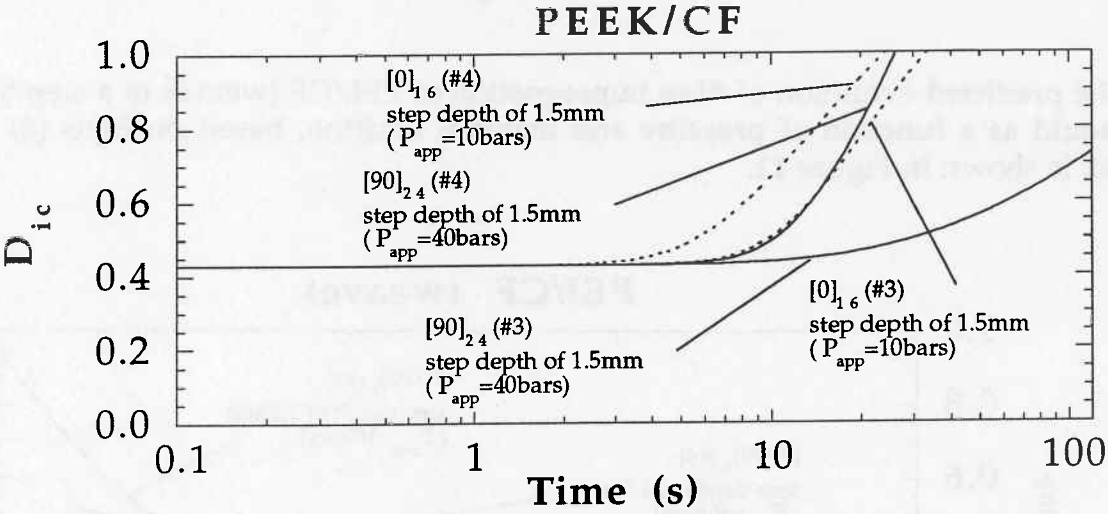

Figure 10: Evolution of D_{ic} for PEEK/CF as a function of time, material location (#3: inside or #4: outside of the central section), fibre orientation and pressure.

For non-preconsolidated PEI/CF (weave), the consolidation is limited by the fibre impregnation. In a previous study, an equation to predict the evolution of the degree of fibre impregnation for PEI/CF (weave) was derived [3,7]:

$$\frac{d^2(t)}{8} - \frac{d_o d(t)}{4} + \frac{d_o d_i}{4} - \frac{d_i^2}{8} = \frac{K_p}{\eta_{PEI/CF(weave)}}(\phi P_{app} - P_{atm})t \qquad (5)$$

where d_o is the thickness of the tow, d_i is the initial thickness of the non-impregnated zone, $d(t)$ is the thickness of the non-impregnated zone at time t, K_p is the fibre bed permeability, P_{atm} is the atmospheric pressure, P_{app} is the applied pressure and ϕ is the ratio between the applied pressure and the pressure transmitted to the resin. The value of the constants required by Eqn (5) are listed in Table 3.

Table 3: Value of the constants K_p, d_i, d_o and ϕ [3,7].

K_p (m^2)	d_i (µm)	d_o (µm)	ϕ (-)
$1.1 \cdot 10^{-13}$	50	79	0.73

The degree of fibre impregnation is defined as:

$$D_{imp}(t) = \frac{d_o - d(t)}{d_o} \tag{6}$$

The predicted evolution of fibre impregnation of PEI/CF (weave) in a step taper mould as a function of pressure and material location, based on Eqns (5) and (6), is shown in Figure 11.

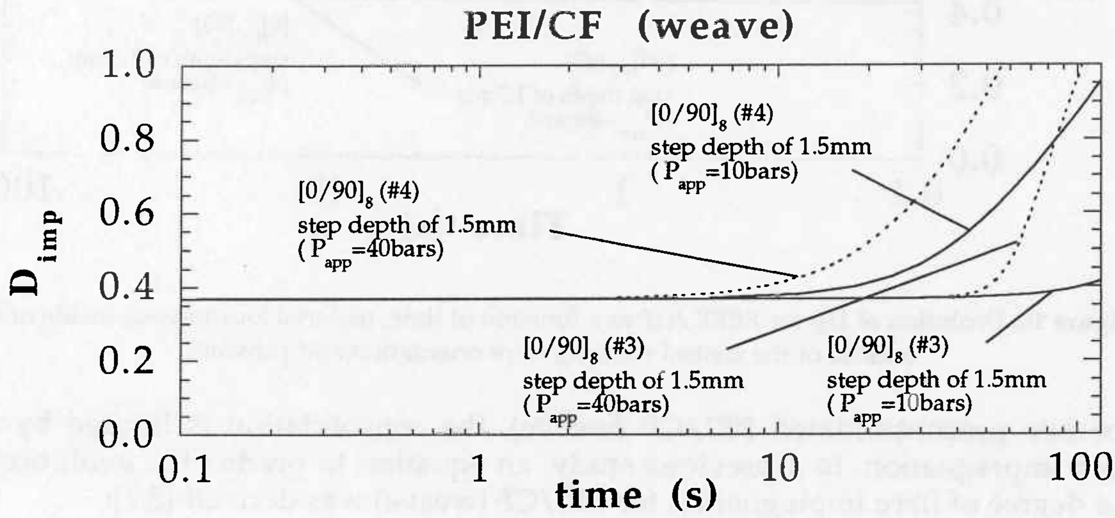

Figure 11: Evolution of D_{imp} for PEI/CF (weave) as a function of time, material location (#3: inside or #4: outside of the step), fibre orientation and pressure.

Material inside the central section of the mould would not consolidate at an applied pressure of 10 bars. For an applied pressure of 40 bars, the whole part was fully consolidated regardless of the fibre orientation, step depth or preform thickness. Consequently, for both materials, the predictions confirm that an inhomogeneous pressure distribution within a part induces only partial consolidation. However, pressure gradients may be prevented by correct mould and preform design, which depends mainly on the material configuration. For unidirectional fibre-reinforced PEEK, the occurrence of pressure gradients depends on the fibre orientation of the preform relative to the mould geometry, whereas for woven fibre-reinforced PEI it depends on the applied pressure.

4. CONCLUSIONS

The consolidation of pressformed PEEK/CF and PEI/CF (weave) parts has been shown to depend on pressure gradients resulting from the part geometry and material configuration. Such gradients especially occur during the forming of complex geometries; they have been generated experimentally by using a step-tapered mould.

The pressure efficiency (PE) during the forming of step-tapered PEEK/CF and PEI/CF (weave) depends on the preform configuration and mould geometry. For PEEK/CF, a low PE was achieved for a fibre orientation of 90°, but a high PE was obtained at other fibre orientations (0° and 45°). For PEI/CF (weave), low and high PE were achieved for applied pressures of 10 and 40 bars, respectively. For both materials neither the preform thickness nor the step depth had a strong influence on the magnitude of the PE.

Observations of the microstructure and modelling of the consolidation behaviour of these step-tapered parts have shown that high pressure gradients (low PE) prevent consolidation in the step zone. In this study, for PEEK/CF and PEI/CF (weave), the degree of consolidation was limited by the degree of intimate contact and fibre impregnation, respectively.

For both materials fibre buckling was observed for certain fibre orientations.

5. ACKNOWLEDGEMENTS

This work was funded by the Swiss Federal Office for Education and Science (OFES) and by the Swiss Commission for the Encouragement of Scientific Research and Science (CERS). It was carried out as a part of the European Commission BRITE-EURAM II project number BE-5092.

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