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Self-reinforced prepregs and laminates of a poly(phenylene oxide)–polystyrene alloy with a liquid crystalline polymer

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Extruded prepregs of a blend of a poly(phenylene oxide) (PPO)–polystyrene (PS) alloy with a liquid crystalline polymer (LCP) have been prepared for various concentrations and with different extension ratios. These prepregs have then been compression moulded into laminates with two orientations of the prepregs in the laminate. In one case, 16 prepreg sheets have been placed in unidirectional orientation. In the other, the 16 sheets have been placed in such a way that each subsequent sheet was at a 45° angle with respect to the preceding sheet, thereby yielding a quasi-isotropic orientation. The laminates have been prepared for each extension ratio with different reduction ratios. The unidirectional laminates have been tested for the tensile strength and secant modulus in the machine direction and the transverse direction. The quasi-isotropic laminates have also been tested for mechanical properties. The mechanical properties of the laminates have been compared to the values obtained for injection moulded blends of the same materials. Prediction of the mechanical properties of the quasi-isotropic laminates have been carried out based on the values obtained for the unidirectional laminates tested in the machine and transverse directions. The mechanical properties of the quasi-isotropic laminates with high extension and reduction ratios are either comparable to or higher than those obtained for the injection moulded blends of the same concentration tested in the machine direction. The morphologies of the prepreg sheets with various extension ratios have been studied in order to determine the nature of the LCP fibres in the prepregs. The morphologies of the quasi-isotropic laminates with various LCP concentrations have also been studied in order to determine the effect of orientation of the LCP fibres in the laminates. The studies have shown that this novel processing technique enables the preparation of self-reinforced laminates of LCP–thermoplastic blends, with controlled anisotropy and enhanced mechanical properties, when compared with the corresponding injection moulded blends.

(Keywords: LCP–thermoplastic blend; prepregs; laminates)

INTRODUCTION

The blending of thermotropic liquid crystalline polymers (LCPs) with thermoplastics has been widely studied[1–11]. It has been known that there are two distinct advantages in using a LCP as a reinforcing agent for the thermoplastics. The LCP is known to form *in situ* reinforcing fibres when blended with the thermoplastic. This is due to the inherent nature of the LCP to form elongated rod-like structures in the nematic phase during the processing. Thus, the use of a LCP helps circumvent processing problems while using other reinforcing agents such as glass fibres, because of the *in situ* fibre formation during processing. The other advantage of using the LCP is that it reduces the viscosity of the blend during the processing[12]. The LCP thus acts as a processing aid, particularly for those thermoplastics that pose significant processing problems owing to their high viscosity.

One of the disadvantages of using a LCP as a reinforcing agent in thermoplastics is the anisotropy of the mechanical properties of the products made from the blends. The LCP forms long fibres in the machine direction due to the shear and extensional flow at the die. Thus the blends have very high mechanical properties in the machine direction, while the properties in the transverse direction are generally poor.

To resolve the problems concerning anisotropy of the thermoplastic–LCP and LCP–LCP products, a technology for manufacturing composite laminates was proposed[13–16]. Recently, some studies have also been published relevant to this technology[17–25]. This present effort is to further advance the processing technology so as to overcome the problem of the anisotropy of the PPO–PS/LCP blends. Prepregs of the blend have been prepared at a temperature above the glass transition temperature (T_g) of the thermoplastic. Laminates of these blends, with multiple prepreg layers have been prepared at a temperature above the T_g of the thermoplastic matrix and below the melting point of the LCP. This helps in conserving the nature and orientation of the fibres of the LCP while allowing a flow of the thermoplastic among the various layers to produce a solid laminate.

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MATERIALS AND METHODS OF INVESTIGATION

The materials used in this study were alloys of PPO and PS (Noryl 731, General Electric) and a thermotropic LCP based on hydroxy benzoic acid (HBA)/hydroxy napthoic acid (HNA) (Vectra A950, Hoechst Celanese). Prepregs and laminates of the PPO–PS/LCP with four different concentrations of the LCP (10, 25, 50 and 75 wt%) were prepared.

Prepreg preparation

The prepregs were prepared by using a coat-hanger die attached to a static mixer, which was, in turn, connected to a single-screw extruder. This equipment was described in ref. 24. The coat-hanger die was used to produce the prepregs in the form of thin sheets of sufficient width to obtain uniform and suitably large laminates for testing. The coat-hanger die was set to a lip gap of 2 mm. The width of the sheets that were made was ~5 cm. The pressure developed was measured at the exit of the extruder. The typical conditions maintained for the production of the prepregs are given in *Table 1*. The speed of the extruder was set at 50 rpm and the speed of the take-up was varied in order to produce sheets of different extension ratios. The extension ratio is defined as the ratio of the speed of the take-up to the speed of extrusion. Sheets with four different extension ratios were prepared for each of the blend concentrations. It was found that the pressure increased with a decrease in the LCP concentration in the blend, indicating an easier flow due to a reduction in the viscosity, because of the addition of the LCP to the system[26].

Preparation of laminates from the prepregs

The prepregs were cut to a size of ~75 × 125 mm, with each laminate composed of 16 such sheets. The lamination was performed in a computer-controlled compression moulding press (Tetrahedron, MTP 24). The platen size of the press was 600 × 600 mm. The maximum force that could be applied by the platens was ~150 tons (~1500 kN). The closing system was hydraulic and the accuracy was ±1% of the maximum force. A maximum temperature of 482°C (900°F) could be applied by means of planar heaters mounted on the platens. The temperature and pressure applied on the sample could be measured by means of thermocouples and pressure transducers (Dynisco) that could withstand high temperatures. The sensors were flush mounted so as to make a contact with the laminate surface during the consolidation process. The platens were cooled by water circulation at a rate of 10°C min^{-1}.

The entire operation of the press was computer controlled. The heating could be carried out in stages. The time and temperature of each step could be preset, in order to ensure uniform heating of the platens. The pressure and temperature data from measurements by the sensors were obtained using a data management system based on a Metra-Byte acquisition (DAS8 and EXP16) which was hooked on to an IBM PC computer. The data processing was performed by using a Notebook software program.

A total of 16 sheets of prepregs were laminated in two orientations. In the first case, all of the 16 sheets were placed parallel to each other along the machine direction. The unidirectional laminates produced by this orientation were used for mechanical testing in the machine and transverse directions. In the other case, the 16 prepreg sheets were placed in such a sequence that each subsequent layer was at an angle of 45° with respect to the previous layer. This orientation made sure that the fibres in the laminate were distributed uniformly in all directions. The laminates thus generated tended to have a quasi-isotropy. These laminates were also tested for their mechanical properties.

The press was heated to 215°C (420°F). This operating temperature was selected to ensure that the LCP did not melt, but the thermoplastic matrix, however, could flow between the various layers to cause the formation of a good laminate. The stack of prepregs were placed in the centre of the platen with a direct contact of the sensors with the stack. Sheets of both Kapton film and cloth were placed on the top and bottom of the stack to protect the sensitive pressure transducer and also facilitate the removal of the laminate after the process. Four different spacers of thickness 1.01, 1.78, 2.29 and 2.79 mm (0.04, 0.07, 0.09 and 0.11 in) were used to produce laminates with four different reduction ratios. The reduction ratio is defined as the ratio of the sum of the thicknesses of all of the 16 prepreg laminated sheets to the final thickness of the laminate. The spacers governed the extent to which

Table 1 Typical conditions for prepreg sheet production[a,b]

Sample	Speed of the extruder (rpm)	Speed of the take-up (fpm)	Extension ratio
1	50	50	6.9
2	50	70	9.6
3	50	90	12.4
4	50	120	16.6
Sample	**Blend ratio of PPO–PS/LCP**	**Pressure developed (MPa)**	**Extension ratio**
1	10/90	3.73	16.6
2	25/75	3.99	16.6
3	50/50	4.21	16.6
4	75/25	4.48	16.6

[a] Temperature settings: single-screw extruder, all zones at 530°F (273°C); static mixer, all zones at 525°F (273°C)
[b] Distance between take-up drum and die, 0.25 in (0.635 cm)

each stack of prepregs was compressed. It was seen that with an increase in the extension ratio, the thickness of the prepregs decreased. This led to the use of a smaller set of spacers during the lamination process for prepregs of a high extension ratio, in order to ensure proper closing of the platens and to eventually obtaining a good laminate.

The force applied by the platens on the prepreg sheets was set to 35.6 kN and this was maintained constant throughout the process. The optimum force and temperature were selected based on trial runs. The criterion for selection was that the laminate produced should be hard and even, with neither delamination nor significant variation in the thickness of different parts of the final product. The sheets were compressed for a period of 15 min at this temperature and pressure. The platens were then cooled down by water circulation to around room temperature before the removal of the laminate. The procedure was repeated for four concentrations of LCP in the blend. For each concentration, laminates were prepared from prepregs obtained at four different extension ratios. For each extension ratio, a number of laminates were prepared with different (four) reduction ratios.

Figure 1 shows the typical data output generated by the pressure and temperature sensors during the consolidation process. It can be seen that the pressure builds up drastically, immediately after the closing of the platens, and then comes down to a steady-state value during the consolidation process. When the cooling is initiated with the platens still being closed, the pressure rises again due to thermal shock and after that it gradually relaxes to a zero value. The observed process is similar to that found in the consolidation process of thermoplastic composites with long carbon fibres[27]. However, the decay of the pressure in this present case of a thermoplastic/LCP laminate is faster.

Mechanical testing of the laminates and prepregs

Sample preparation. The prepregs and laminates that were produced were cut to a dumb-bell shape with a die conforming to a standard mini tensile bar (MTB) size of 0.635 × 0.0031 × 0.0015 m using a Router/Saber saw mill (American Vermont, No 23466, with a motor speed of 25 000 rpm). At least five samples were cut from each laminate and these were used for mechanical testing. In the case of unidirectional laminates, the samples were cut in both machine and transverse directions. The dimensions of each of the samples were carefully recorded in order to ensure elimination of any error in calculations due to dimensional changes among the samples. Single sheets of the prepregs that were used for lamination were also tested for their mechanical properties so as to find the extent of loss of strength due to the consolidation process.

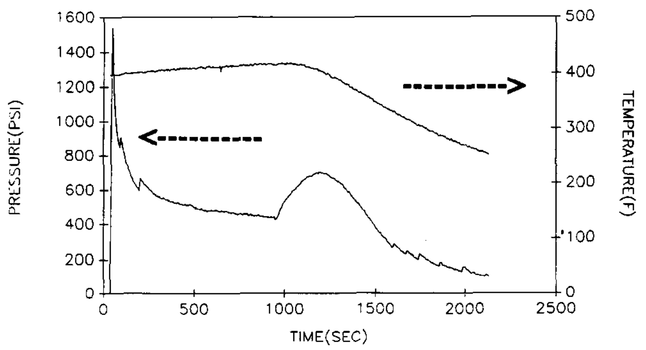


Figure 1 Typical pressure and temperature traces produced during consolidation of a 50/50 PPO–PS/LCP quasi-isotropic laminate with a reduction ratio of 1.8; prepregs used were prepared with an extension ratio of 16.6

Mechanical testing. The mechanical properties of the laminates were obtained by using a MTS mechanical tester capable of a cyclic operation (Model 305.03, serial number 62). The MTBs that were cut as described above had smooth, glass-like surfaces; the MTS tester, with hydraulic clamps, was found to hold these types of sample better (with a pressure of 2–3 MPa). A triangular mode of operation, with a very slow head movement (0.01–0.11 Hz frequency), converted the fatigue tester into a mechanical properties tester, since the MTBs tended to break during the extension period of the cycle. Moreover, the compression period of the cycle placed the head in the exact same starting position, thus contributing to the consistency of the tests. The results were obtained through a microprocessor (MTS micro console 458.20), and the peak tensile strength could be directly read from this. A chart recorder was used to record the movement of the head, and the readings obtained were used to calculate the secant modulus and elongation to break. Secant modulus is defined as the modulus of the sample at 1% elongation. The cross-head speed was set at 5 mm min^{-1}, while the chart speed was set at 20 cm min^{-1}. At least five samples were tested for each laminate, with the tests also being performed for single sheets of prepregs. In these cases, the pressure on the clamps was reduced to less than 1 MPa in order to ensure that the sheets did not crack due to the clamp pressure during the testing.

Morphology of prepregs and laminates. The morphologies of the prepreg sheets of 25% LCP concentration for different extension ratios were studied in order to observe the nature of the LCP fibres in the single sheet. The morphologies of the quasi-isotropic laminates for different LCP concentrations were also studied to reveal the nature of the LCP fibres in the laminate after consolidation. The morphology was examined by using a scanning electron microscope (SEM ISI SX-40). Small strips of single prepreg sheets with different LCP concentrations were immersed in liquid nitrogen and flexed to fracture. The samples were mounted, with the cross-section exposed, on sample holders using a quick-drying cyanoacrylate and then coated with a gold–palladium alloy using a Polaron SEM coating machine; the coating time was fixed at 60 s. The scanning electron microscope pictures were used to observe the orientation of the LCP fibres in the sheets.

RESULTS AND DISCUSSION

Figure 2 shows a typical stress–strain curve for a MTB cut from a PPO–PS/LCP quasi-isotropic laminate with a 50% LCP concentration. The stress was calculated from the force on the moving head of the clamps while the strain was obtained from the chart displacement. The laminates typically show a brittle behaviour as indicated by the figure.

Figure 3 shows the tensile strength and secant modulus as a function of the reduction ratio for various extension ratios of PPO–PS/LCP unidirectional laminates containing 10% LCP concentration, tested in the machine (top) and transverse (bottom) directions. It can be seen that the tensile strength and modulus of the laminate tested in the machine direction increases both with the increase in reduction ratio of the laminate and with the increase in extension ratio of the prepregs. An increase in the extension ratio causes a higher LCP orientation in the machine direction and thus stronger LCP fibres are formed *in situ* during the extrusion and stretching of the prepreg. This would account for the increases in the tensile strength and modulus of the laminates with the increase in the extension ratio of the prepregs. The increases in the tensile strength and modulus with reduction ratio can be attributed to an additional orientation of the LCP fibres during the consolidation process. This could be the result of shear and elongational forces imposed during this process. Two conflicting phenomena are thought to occur during the consolidation process. One is the release of orientation due to the heating (annealing) and the other is the orientation build-up due to the viscous forces acting during the process[23]. These two competing phenomena dictate the final orientation of the fibres and consequently the strength and modulus of the laminate as a function of the reduction ratio. In the case of these PPO–PS/LCP blends, the temperature of consolidation is much lower than the melting point of the LCP and thus the release of orientation due to heating is less than the orientation build-up due to consolidation. This accounts for the increases in tensile strength and modulus of the laminate with the increase in reduction ratio.

The ordinate of *Figure 3* gives the tensile strength and modulus for the injection moulded sample of the blend of PPO–PS/LCP with 10% LCP concentration (taken from ref. 26), and also the values for the single prepreg sheets of different extension ratios tested in the machine direction. It can be seen that the values of the laminates

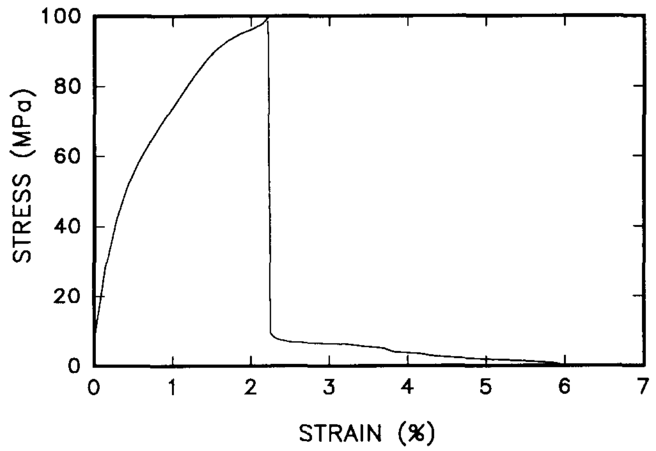


Figure 2 Stress–strain curve for a mini tensile bar cut from a unidirectional laminate of 50/50 PPO–PS/LCP prepared with an extension ratio of 16.6 and a reduction ratio of 1.8

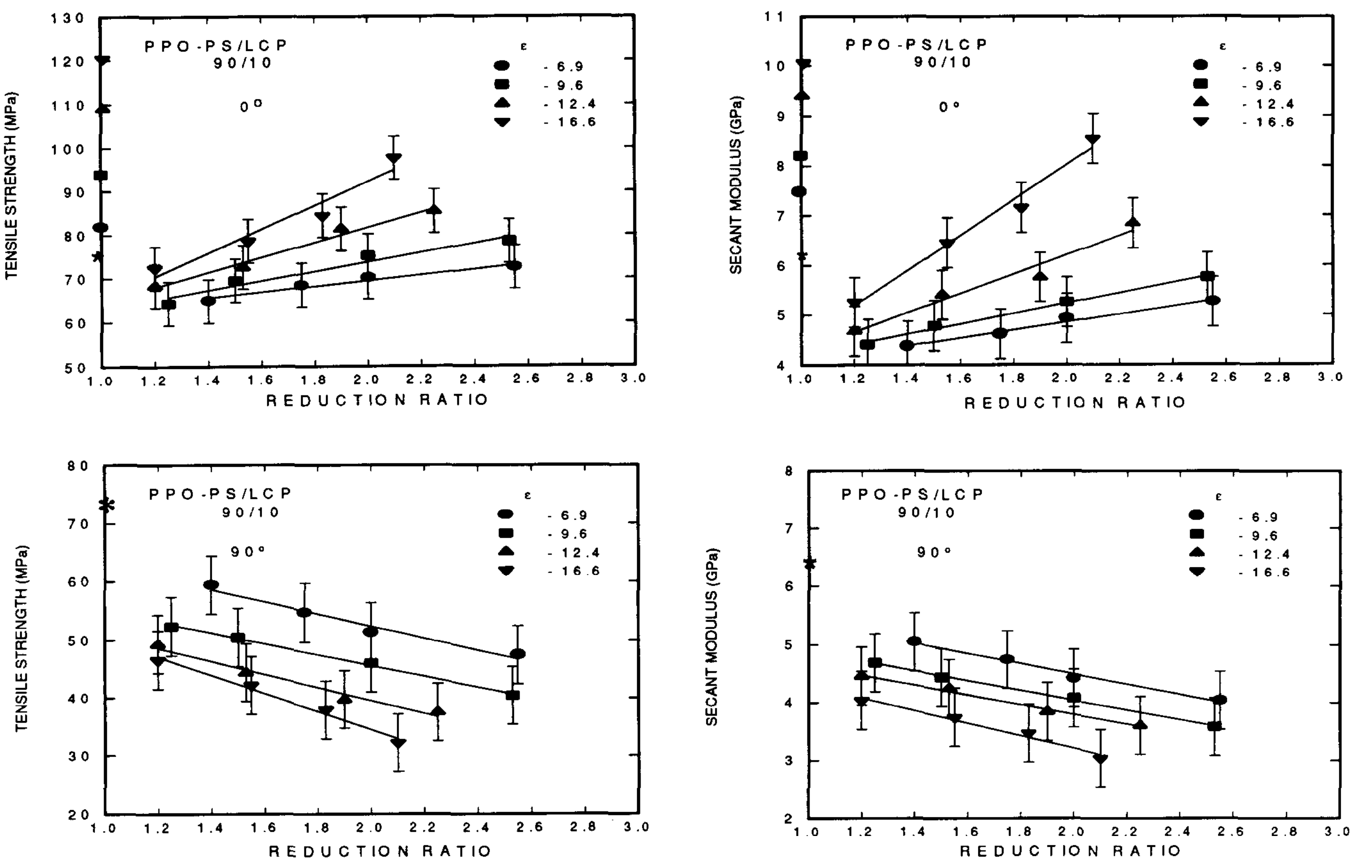


Figure 3 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 90/10 PPO–PS/LCP unidirectional laminates, tested in the machine (top) and transverse (bottom) directions. The symbols on the *y*-axis are the values for the single prepreg sheets; the asterisk represents the value obtained for injection moulded blends in the flow direction[26]

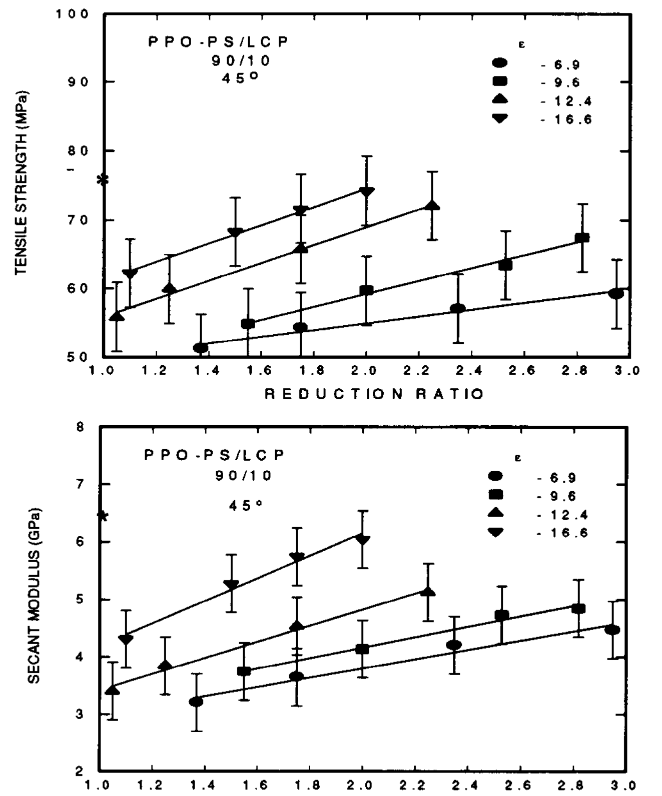


Figure 4 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 90/10 PPO–PS/LCP quasi-isotropic laminates; the asterisk represents the value obtained for injection moulded blends in the flow direction[26]

are lower than those of the corresponding prepreg sheets. This can be attributed to the loss of orientation of the PPO–PS matrix due to consolidation. It can be seen that the tensile strength and modulus of the laminates are comparable to, or higher than, those of injection moulded samples, particularly at high extension and reduction ratios.

Figure 3 (bottom) shows the tensile strength and secant modulus as a function of reduction ratio of various extension ratios for PPO–PS/LCP unidirectional laminates containing 10% LCP tested in the transverse direction. The figure shows that the tensile strength and modulus decrease with increases in the extension and reduction ratios. As the LCP fibres get more oriented in the machine direction, the strength and modulus in the transverse direction are expected to decrease due to the anisotropy of the properties of the thermoplastic–LCP blends.

Figure 4 shows the tensile strength (top) and modulus (bottom) of PPO–PS/LCP quasi-isotropic laminates with 10% LCP concentration. In this case also, the tensile strength and modulus are found to increase with the increases in extension and reduction ratios. The ordinate shows the value obtained for an injection moulded blend of the same concentration, tested in the machine direction, taken from ref. 26. It can be seen that the values obtained for the laminates with high extension and reduction ratios are quite close to that found for the injection moulded blend. Thus, the strength and modulus of the laminate are not compromised to any great extent due to the quasi-isotropic orientation of the prepregs for laminates of high extension and reduction ratios. This

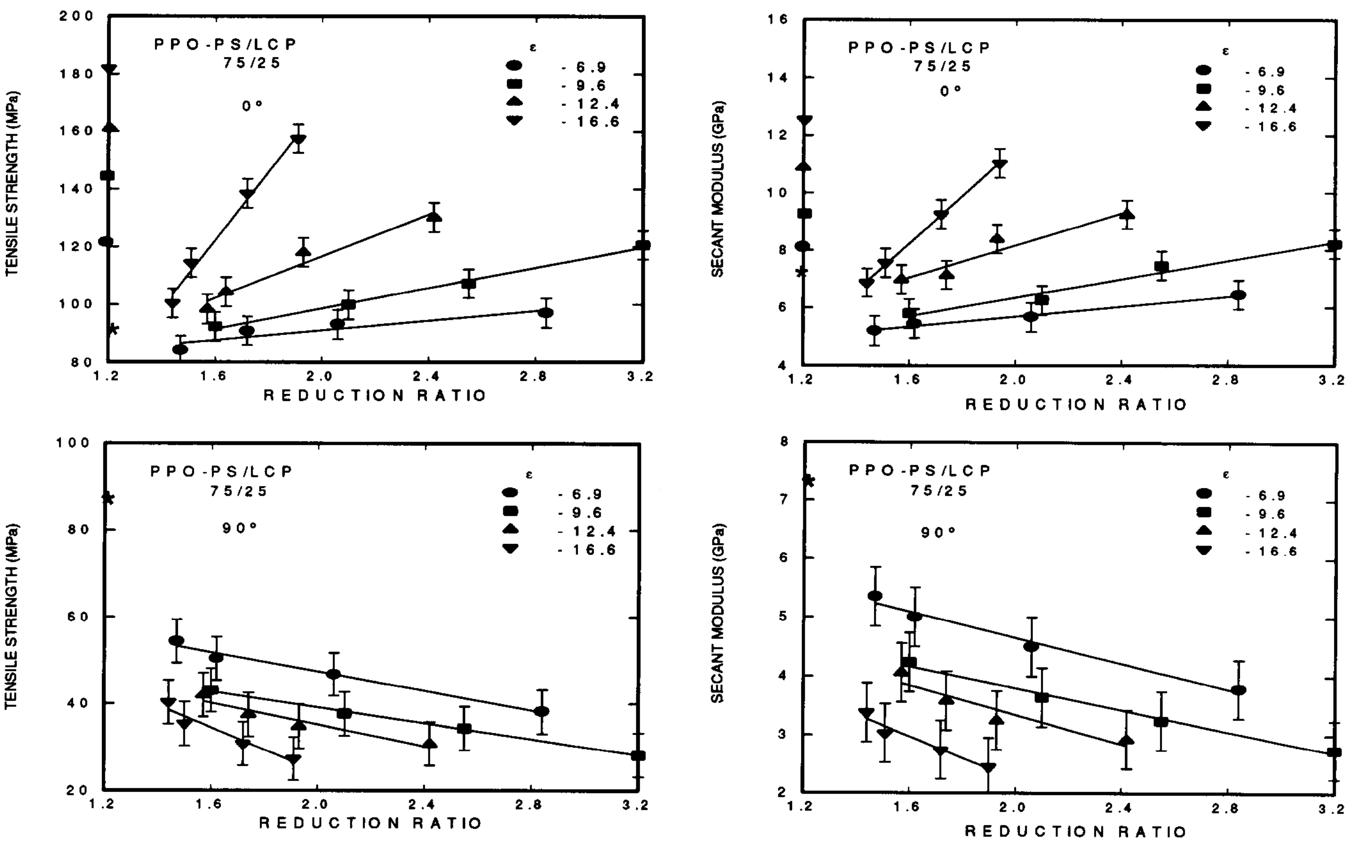


Figure 5 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 75/25 PPO–PS/LCP unidirectional laminates, tested in the machine (top) and transverse (bottom) directions. The symbols on the *y*-axis are the values for the single prepreg sheets; the asterisk represents the value obtained for the injection moulded blends in the flow direction[26]

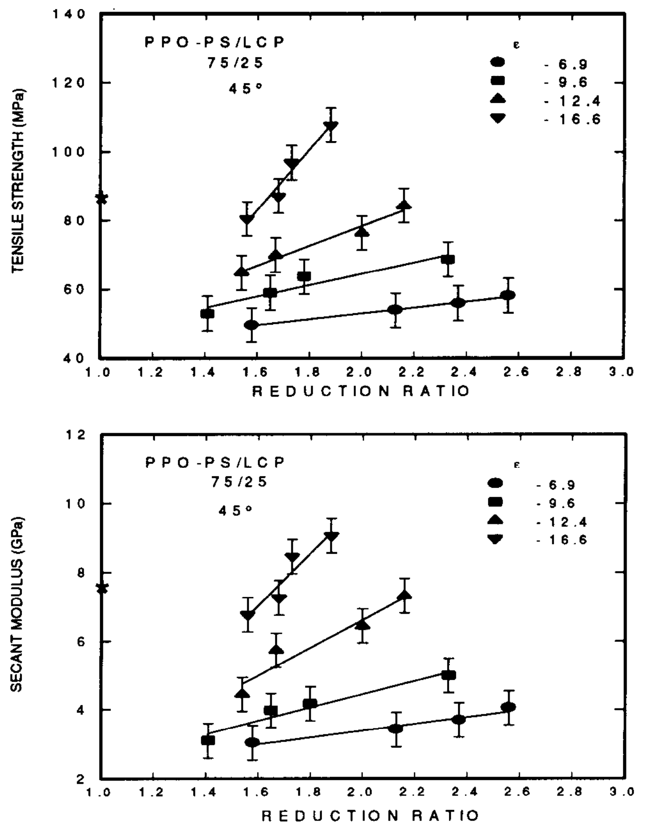


Figure 6 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 75/25 PPO–PS/LCP quasi-isotropic laminates; the asterisk represents the value obtained for injection moulded blends in the flow direction[26]

result is of importance since a certain amount of isotropy has been established without a loss of a significant amount of strength.

Figure 5 shows the tensile strength and secant modulus of PPO–PS/LCP unidirectional laminates with 25% LCP concentration, tested in the machine (top) and transverse (bottom) directions, as a function of the reduction ratio for various extension ratios. The tensile strength and modulus both increase with an increase in the extension and reduction ratios, very similar to the laminates with 10% LCP concentration. The ordinates in this figure also show the values of the tensile strength and modulus of injection moulded samples of the same concentration[26], and also those of the single prepreg sheets tested in the machine direction. The tensile strength and modulus of the laminates are higher than those obtained for the injection moulded sample at high extension ratios, even at low reduction ratios. This can be attributed to a higher amount of LCP fibres oriented in the machine direction. It appears that the contribution of the stretching conditions at the coat-hanger die exit, along with the viscous flow during consolidation, are more favourable for LCP fibre orientation than the shear and elongational flow occurring during injection moulding. As in the previous case, the prepregs have a higher tensile strength than the laminates, and this is attributed to some loss of orientation due to annealing during the consolidation process. As expected, the tensile strength and modulus both decrease with increases in the extension and reduction ratios for samples with orientation in the transverse direction.

Figure 6 shows the tensile strength and secant modulus

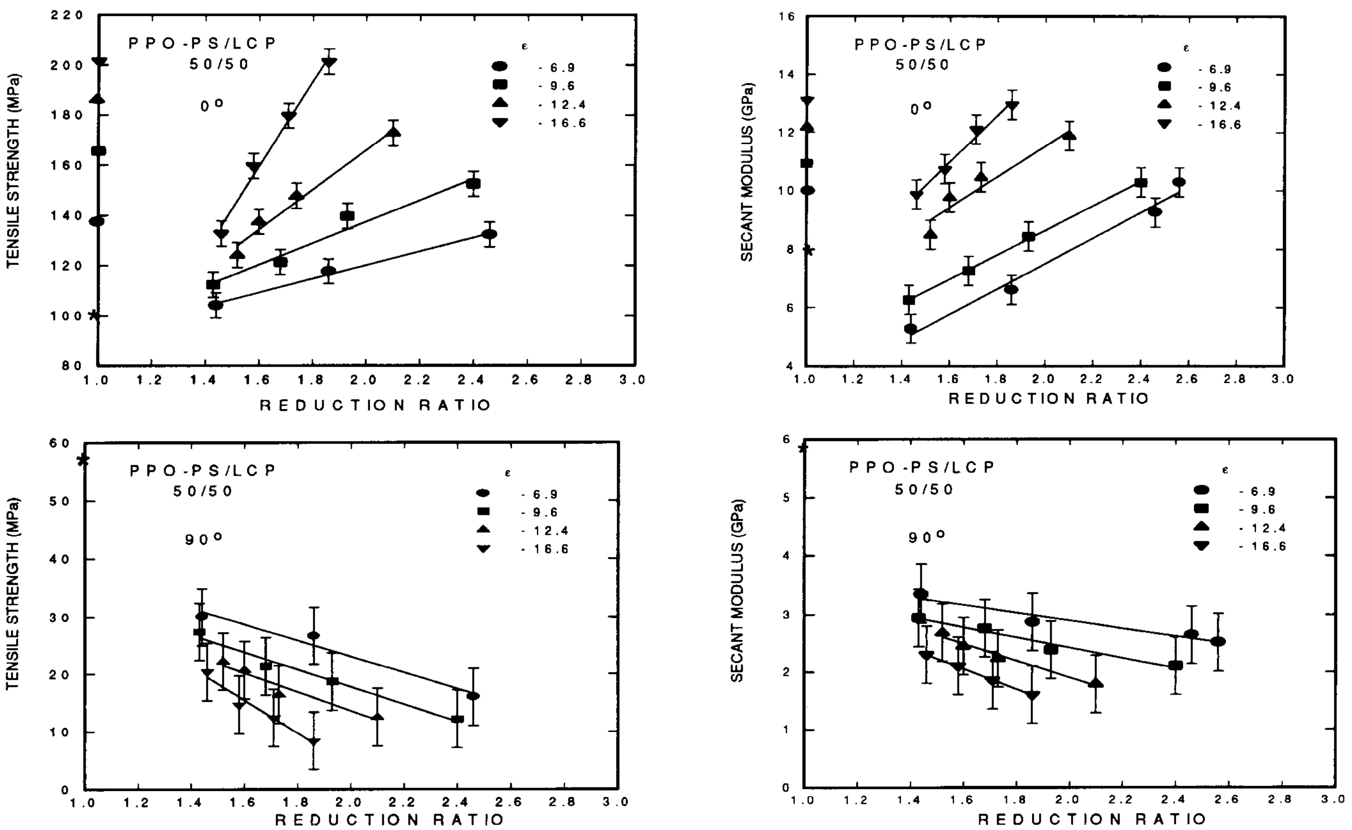


Figure 7 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 50/50 PPO–PS/LCP unidirectional laminates, tested in the machine (top) and transverse (bottom) directions. The symbols on the y-axis are the values for the single prepreg sheets; the asterisk represents the value obtained for injection moulded blends in the flow direction[26]

of PPO–PS/LCP quasi-isotropic laminates with 25% LCP concentration, as a function of the reduction ratio, for various extension ratios. The interesting feature in this figure is the proximity of the tensile strength and modulus values of the laminates to that of the injection moulded sample. These values are closer than in the case of the 10% LCP quasi-isotropic laminates. In this case too, the tensile strength and modulus both increase with increases in the reduction and extension ratios.

Figure 7 gives the variations in the tensile strength and secant modulus of the 50% LCP unidirectional laminates, tested in the machine (top) and transverse (bottom) directions. The trends observed for the 50% laminates are very similar to those of the 25% LCP blends. However, in the case of the 50% LCP laminates, the values of the tensile strength and modulus for the laminates are even higher than those obtained for the corresponding injection moulded samples[26], particularly at high extension and reduction ratios. Moreover, at this LCP concentration, the tensile strength and modulus of the unidirectional laminate at a high extension and reduction ratio become close to those of the single prepreg sheets, indicated on the ordinate of *Figure 7* (top). The values of the tensile strength and modulus for 50% LCP unidirectional laminates tested in the transverse direction are lower than the values obtained for 25% LCP unidirectional laminates tested in the transverse direction. This shows that as the LCP concentration increases, the tensile strength and modulus in the transverse direction decrease significantly, owing to the increase in anisotropy of the unidirectional laminates, because of the presence of LCP fibres which are predominantly oriented in the machine direction.

Figure 8 gives the tensile strength (top) and secant modulus (bottom) of the PPO–PS/LCP quasi-isotropic laminates with 50% LCP concentration. It can be seen that the tensile strength and modulus of the laminates with high extension and reduction ratios may attain a higher tensile strength than that of the injection moulded blend with the same concentration taken from ref. 26 (indicated on the ordinates). As in the case of the 10% and 25% LCP laminates, the tensile strength and modulus of the quasi-isotropic laminates increase with an increase in the reduction and extension ratios.

Figure 9 gives the tensile strength and secant modulus of the 75% LCP unidirectional laminates tested in the machine direction (top) and quasi-isotropic laminates (bottom), as a function of the reduction ratio, for various extension ratios. It was found that the tensile strength and modulus of the unidirectional laminates could not be obtained in the transverse direction because the samples were too brittle and the strength was too low to be measured accurately. This was due to the increase in anisotropy of the unidirectional laminate with the increase in LCP concentration. It can be seen that the tensile strength and modulus of the unidirectional laminates of even low reduction ratios, at high extension ratios, are comparable with or higher than that of the injection moulded sample tested in the machine direction and shown on the ordinates of *Figure 9*. This data point was taken from ref. 26, for the same LCP concentration. It can also be seen that both the tensile strength and modulus of the unidirectional laminates obtained at a high reduction ratio are close to those of the single prepreg sheets indicated on the ordinates of *Figure 9* (top). The tensile strength of the quasi-isotropic laminates with high extension and reduction ratios are comparable with that of the injection moulded blend. The secant moduli of the unidirectional laminates tested in the machine direction are far greater than the modulus of the injection moulded blend also tested in the machine direction[26] and indicated on the ordinate. In fact, the secant moduli of the quasi-isotropic laminates obtained at high extension and reduction ratios are comparable to or higher than those of the injection moulded blends tested in the machine direction and shown in *Figure 9* (bottom). Thus, the data obtained indicate that at higher concentrations of LCP, quasi-isotropic laminates can be prepared with relatively uniform properties without too much of a compromise on the mechanical properties. Therefore, the lamination process thus offers a way to utilize the strength and unique properties of the LCP, when blended with other thermoplastics, while also circumventing the problems associated with anisotropy.

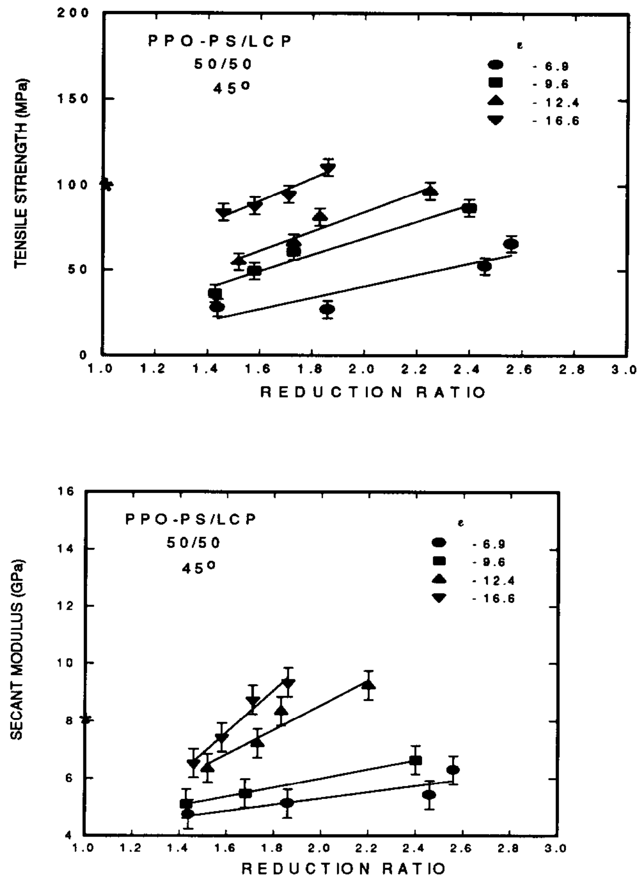


Figure 8 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 50/50 PPO–PS/LCP quasi-isotropic laminates; the asterisk represents the value obtained for injection moulded blends in the flow direction[26]

Predictions for quasi-isotropic laminates

Prediction of the mechanical properties of the quasi-isotropic laminates is based on the assumption that the values of the mechanical properties of the unidirectional laminate in a certain direction vary in an elliptical fashion with the angle of this direction with respect to the fibre orientation. Each layer of the prepreg contributes to the strength of the laminate. The strength of the quasi-

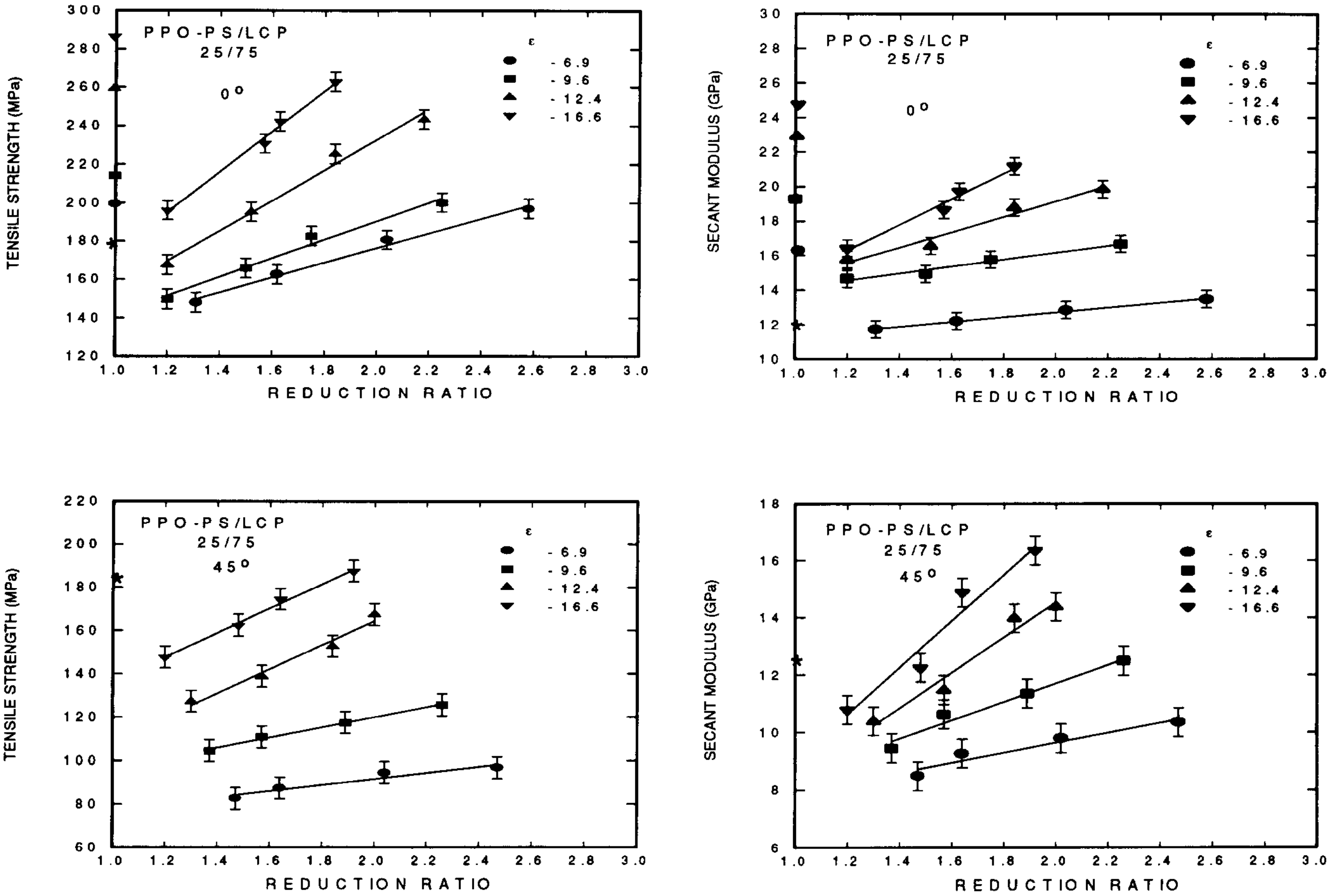


Figure 9 Tensile strength and secant modulus vs. reduction ratio measured for various extension ratios of 25/75 PPO–PS/LCP unidirectional laminates, tested in the machine direction (top), and quasi-isotropic laminates (bottom). The symbols on the y-axis are the values for the single prepreg sheets; the asterisk represents the value for the injection moulded blends in the flow direction[26]

isotropic laminate depends on the orientation of the layers of the prepregs in the lay up. In the case of quasi-isotropic laminates, there is a certain contribution by the prepregs in the parallel and transverse direction towards the strength in a 45° orientation. Thus, the values of the tensile strength and the secant modulus of the quasi-isotropic laminate has been predicted here with a knowledge of the values in the parallel and transverse orientations. *Figure 10* elucidates a basic schematic for prediction of the mechanical properties of the quasi-isotropic laminates from the values for unidirectional laminates tested in the machine and transverse directions. The values of the mechanical properties of the quasi-isotropic laminate are calculated from the following equations:

$$E = \frac{\sum_{i=1}^{n} E_{\mathrm{pl},i} + \sum_{i=1}^{n} E_{\mathrm{pr},i} + \sum_{i=1}^{2n} E_{45,i}}{4n} \tag{1}$$

and

$$E_{45} = \frac{E_{\mathrm{pl}} E_{\mathrm{pr}}}{\sqrt{(E_{\mathrm{pl}} \sin 45°)^2 + (E_{\mathrm{pr}} \cos 45°)^2}} \tag{2}$$

where E_{pl} is the value of the mechanical property contributed by each single sheet in the parallel direction, E_{pr} is the value of the mechanical property contributed by the single sheet in the perpendicular (transverse) direction and E_{45} is the value of the property of the single sheet in the 45° direction. Since a decrease in the properties (annealing) of the unidirectional laminates is seen from *Figure 3*, in comparison with those of the prepregs, the values of E_{pl} and E_{pr} used in the calculations are taken from measurements on the unidirectional laminates. The quasi-isotropic laminates in our studies consist of 16 layers, i.e. 4 sheets in the parallel direction, 4 sheets in the perpendicular direction and 8 sheets in the 45° orientation. Thus, the value of 'n' in equation (1) is equal to 4. The values of the property for the parallel and perpendicular directions are obtained by taking the

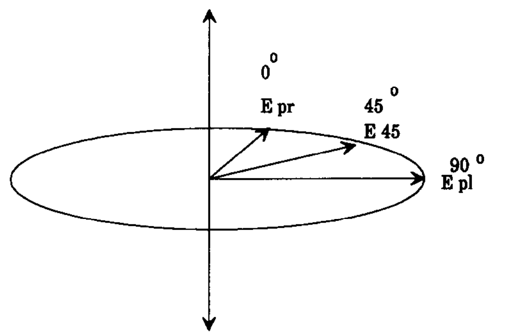


Figure 10 Schematic representation of the ellipse for prediction of the mechanical properties of the quasi-isotropic laminates (marked E_{45}), based on the values obtained for the unidirectional laminates, tested in the machine (marked E_{pl}) and transverse (marked E_{pr}) directions

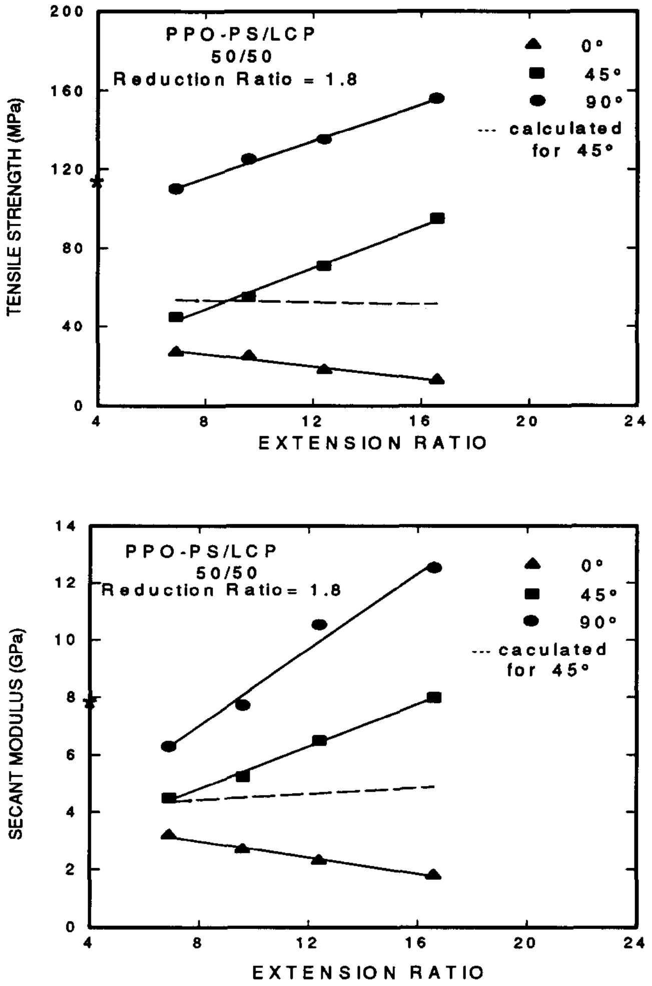


Figure 11 Tensile strength (top) and secant modulus (bottom) vs. extension ratio, for a constant reduction ratio of 1.8, for a 50/50 PPO–PS/LCP unidirectional laminate, tested in the machine and transverse directions, and quasi-isotropic laminates. The dashed line gives the calculated values of the quasi-isotropic laminates, while the asterisk represents the value for the injection moulded blend[26]

value for the unidirectional laminate tested in the machine and transverse directions, and then dividing by 16 to arrive at the contribution of each sheet.

Figure 11 gives the tensile strength (top) and secant modulus (bottom) of the unidirectional laminates with a 50/50 PPO–PS/LCP ratio, tested in the machine and transverse directions, and of quasi-isotropic laminates, as a function of the extension ratio, at a constant reduction ratio. The dotted line indicates the calculated values obtained by equations (1) and (2) for the quasi-isotropic laminates. It can be seen that the experimental values of the tensile strength of the quasi-isotropic laminates are slightly lower than the predicted values at low extension ratios. At high extension ratios, the experimental values are far higher than the predicted values. This increase in the experimentally observed values is attributed to the additional orientation of the LCP fibres due to the consolidation process. At low extension ratios, the probable cause for the tensile strength values being lower than the predicted values is damage to the thicker LCP fibres during the consolidation process. At the highest extension ratio of 16.6, the difference in the predicted and actual values is quite large. At this extension ratio, the LCP fibres, being thinner and longer, are presumed to undergo very little damage during consolidation. In addition, the loss of strength due to fibre damage is very much less when compared to the gain in strength due to the inducement of orientation by the shear and elongational forces acting during the consolidation process. Concerning the predicted and measured secant modulus, *Figure 11* indicates that these values match quite closely at a very low extension ratio, whereas at higher extension ratios, the actual values are much higher than the predicted values. This increase is also attributed, as in the case of tensile strength, to the additional orientation occurring during the consolidation process.

Figure 12 shows the tensile strength (top) and secant modulus (bottom) of various laminates, at a constant extension ratio of 12.4 and a reduction ratio of 1.8, as a function of the LCP concentration in the laminate. The dotted line indicates the predicted values for the quasi-isotropic laminates. It can be seen that the tensile strength of the unidirectional laminates tested in the machine direction, and the quasi-isotropic laminates, both increase with the increase in LCP concentration. The values, as expected, decrease with the increase in the LCP concentration for the unidirectional laminates tested in the

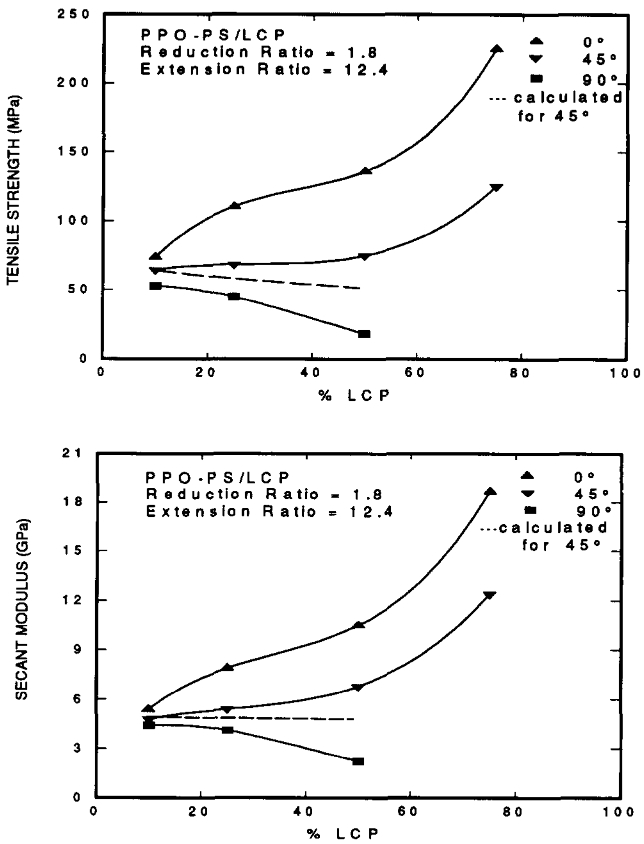


Figure 12 Tensile strength (top) and secant modulus (bottom) vs. the LCP concentration for the PPO–PS/LCP unidirectional laminates, tested in the machine and transverse directions and quasi-isotropic laminates, at a constant extension ratio of 12.4 and a reduction ratio of 1.8; the dashed lines give the calculated values for the quasi-isotropic laminates

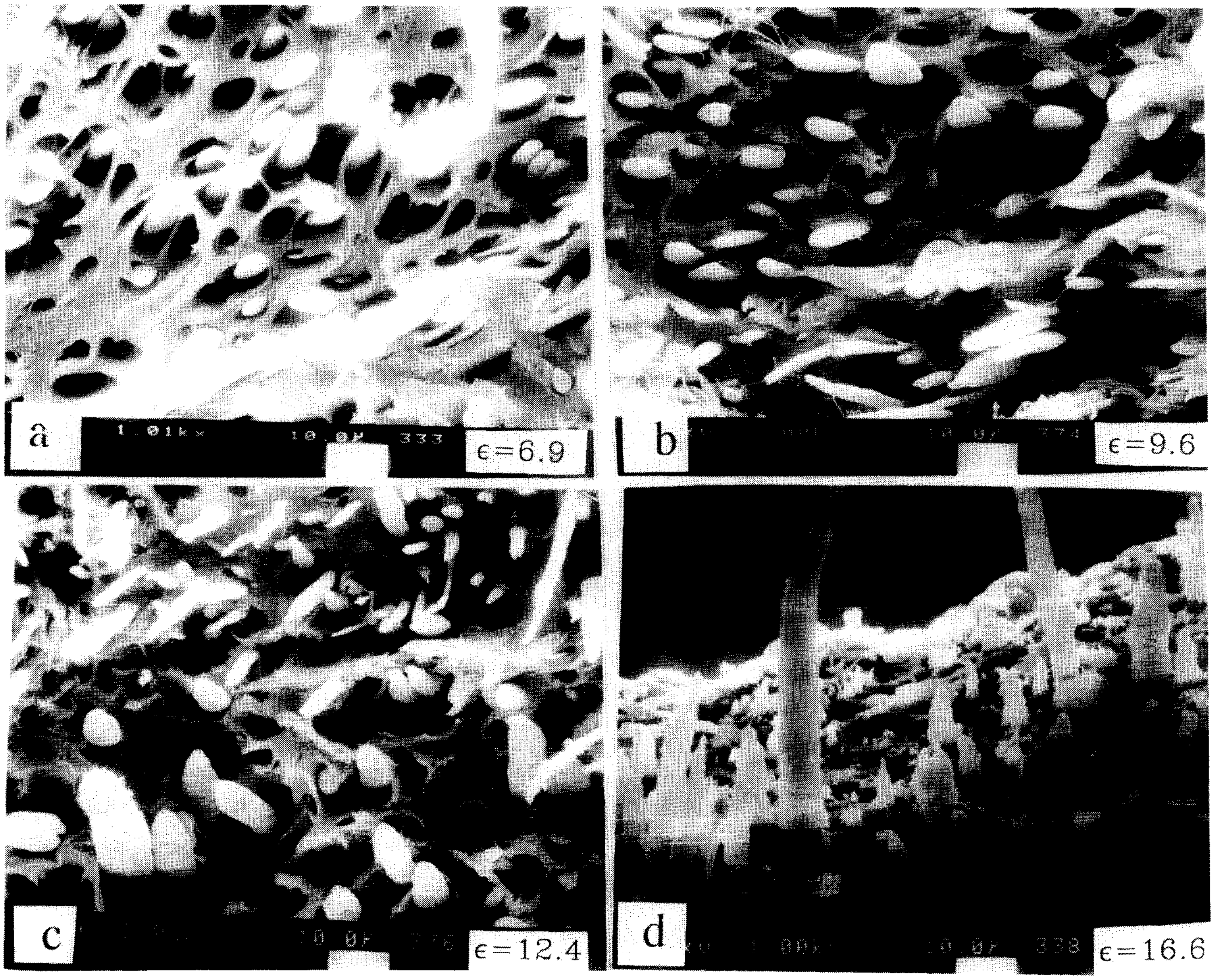


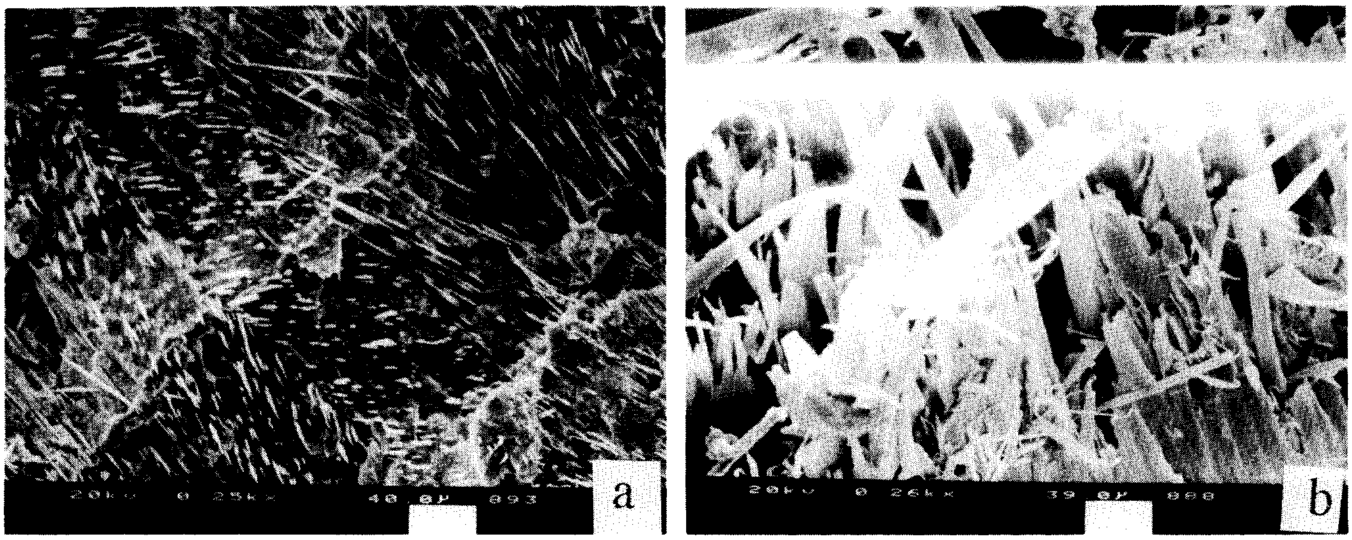
Figure 13 Scanning electron micrographs of 75/25 PPO–PS/LCP prepreg sheets with different extension ratios: (a) 6.9; (b) 9.6; (c) 12.4; (d) 16.6. The samples were quenched in liquid nitrogen and then broken to expose a cross-section of the sheet

transverse direction. The actual values obtained are close to the predicted values, for the quasi-isotropic laminates, at a low LCP concentration. At higher LCP concentrations, the actual values are higher than the predicted values. As the LCP content in the laminate increases, an increase in orientation during the consolidation process is presumed to occur, and this is attributed to the higher actual values. Concerning the secant modulus, *Figure 12* (bottom) shows that the actual secant moduli of the quasi-isotropic laminates is higher than the predicted values at higher LCP concentrations. This increase is also attributed, as in the case of the tensile strength, to the further inducement of orientation of the LCP during the consolidation process.

Morphological properties

Figure 13 shows scanning electron micrographs of prepreg sheets of 75/25 PPO–PS/LCP composition at various extension ratios. At low extension ratios, the LCP component shows an extended globular morphology in the prepreg. The number of voids decreases with the increase in the extension ratio. As the extension ratio increases, the LCP fibres are generated, and these seem to become longer and more slender with further orientation in the machine direction. During the consolidation process, at a particular reduction ratio, it can be expected that the thicker fibres are more susceptible to damage than the thinner fibres. Thus, the morphology of the fibres in the prepregs could explain the significant decrease in the observed mechanical property values of the quasi-isotropic laminates when compared to the predicted values at low extension ratios, as seen in *Figure 11*. At the highest extension ratio of 16.6, the LCP fibres seem to be long, and the matrix is well packed, with less voids. The better packing of the matrix and the higher orientation of the LCP fibres in the prepreg, at high extension ratios, may be the reason for the high mechanical strength of the corresponding laminates.

Figure 14 shows scanning electron micrographs of the quasi-isotropic laminates with 25%, 50% and 75% LCP concentrations. In order to distinguish the layered structure, these micrographs have been obtained at low magnifications. It can be seen from this figure that the quasi-isotropic laminates containing prepreg layers in



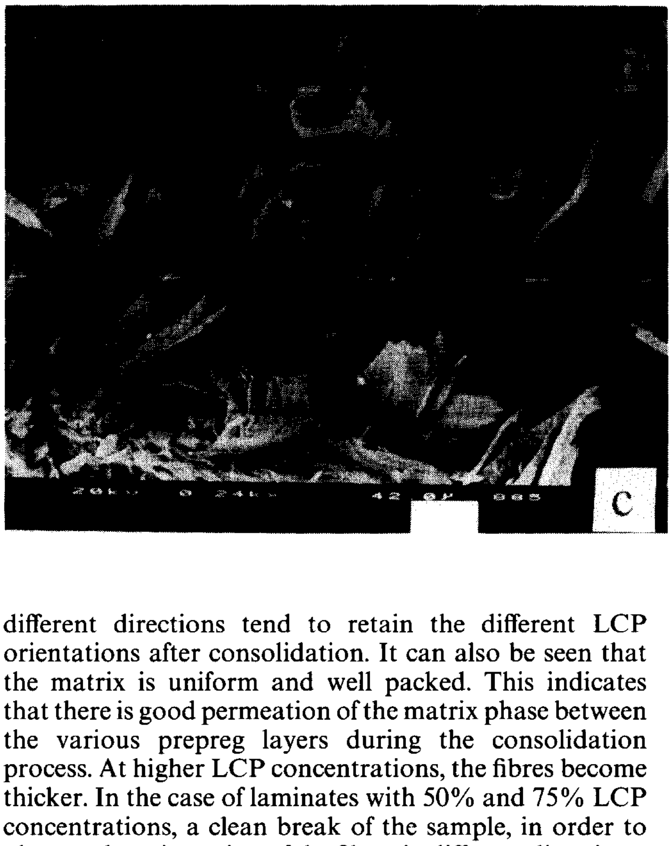


Figure 14 Scanning electron micrographs of quasi-isotropic laminates with different LCP concentrations, obtained at an extension ratio of 12.4 and a reduction ratio of 1.8: (a) 25%; (b) 50%; (c) 75%. The samples were quenched in liquid nitrogen and then broken to expose a cross-section of the sheet

different directions tend to retain the different LCP orientations after consolidation. It can also be seen that the matrix is uniform and well packed. This indicates that there is good permeation of the matrix phase between the various prepreg layers during the consolidation process. At higher LCP concentrations, the fibres become thicker. In the case of laminates with 50% and 75% LCP concentrations, a clean break of the sample, in order to observe the orientation of the fibres in different directions, could not be obtained due to the high strength of the fibres. It can be seen from *Figure 14* that the LCP fibres tend to get thicker and agglomerate into bundles, with the increase in LCP concentration in the laminate. This type of morphology accounts for the high strength of the laminates with high LCP concentrations.

CONCLUSIONS

Self-reinforced PPO–PS/LCP prepregs have been prepared by stretching the sheets obtained from a coat-hanger die attached to an extruder– static mixer set-up. The prepregs have been stretched to various extension ratios. The processing temperature at this stage has been set to facilitate melt processing of both of the components. Stacks of these prepreg sheets have been placed in parallel and quasi-isotropic orientations and consolidated to obtain laminates. The temperature for the lamination process has been selected to be below the melting point of the LCP but above the glass transition temperature of the matrix thermoplastic in order to allow the matrix polymer to flow between the prepreg layers during consolidation. The mechanical properties of these laminates have been measured and compared with injection moulded samples. It has been found that the unidirectional laminates with high extension and reduction ratios tested in the machine direction have better mechanical properties than the injection moulded samples. The laminates with quasi-isotropic orientation of the prepreg layers have been found to have mechanical properties that are either equal to or exceed those obtained for injection moulded samples tested in the machine direction, particularly at high extension and reduction ratios and high LCP concentrations. The values of the mechanical properties of the quasi-isotropic laminates, at high extension ratios, have been found to be much higher than the predicted values based on the values obtained for the unidirectional laminates tested in the machine and transverse directions. This has been attributed to the inducement of orientation during the consolidation process. Morphological studies have indicated the presence of long LCP fibres in the prepregs. These fibres have been found to become more elongated and oriented in the machine direction, with a closer packing of the matrix, as the extension ratio increases. The morphology of the quasi-isotropic laminates indicates that the LCP fibres tend to retain their direction of placement in the lay up, even after consolidation, and this leads to the development of isotropy in these samples. The described technique is therefore found to be a viable method for preparation of self-reinforced laminates using LCP–thermoplastic blends with a controlled anisotropy and enhancement in the mechanical properties, when compared with injection moulded blends.

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