

LOW PRESSURE PROCESSING OF HIGH FIBER CONTENT COMPOSITES

V. Michaud, S. Sequeira Tavares, A.Sigg, S.Lavanchy and J.-A.E. Månson

Ecole Polytechnique Fédérale de Lausanne (EPFL)

Laboratoire de Technologie des Composites et Polymères (LTC)

CH-1015 Lausanne, Switzerland

ABSTRACT: The present work proposes a methodology to establish the process window for void-free carbon fiber-epoxy composites, based on prepregs processed under low pressure. The maximal fiber volume fraction that can be attained for a given pressure was measured from prepreg compaction curves. Three different prepregs, and two different stacking sequences were investigated. The curves all exhibited a very similar behavior, for a given stacking sequence, which provided an upper boundary of achievable volume fraction fiber, for a given pressure differential. The stacking lay-up was shown to exert an influence, as well as the fiber modulus. In addition, the role of the initial resin content and of the consumables was evaluated, to show that with a high initial resin content, resin bleed may allow the part to reach the maximal volume fraction fiber. On the other hand, with limited bleeding, high fiber content may be reached by selecting the initial resin content based on the process window.

KEYWORDS: thermoset composites, fiber bed compressibility, low pressure processing

INTRODUCTION

High fiber content composites are currently often produced by autoclave processing of prepreg plies. Using this technique, composite parts having over 65 vol% carbon fibers and less than 0.5% voids are available, mostly for aeronautic or space applications. The increasing use of carbon-epoxy composites, in particular for large structures, such as aeronautic parts or yacht hulls now leads us to explore the feasibility of processing these materials with the same requirements of fiber and void content using vacuum bag processing only, or limited application of pressure.

When an assembly of prepregs with a given stacking arrangement is processed under a given applied pressure (about 0.9 bar for vacuum-bag processing, up to several bars in an autoclave), the externally applied pressure P_{ext} is supported both by the fiber network and the resin, as follows: $P_{ext} = \sigma_{eff} + P$, where σ_{eff} is the effective stress supported by the fiber network, and P the resin pressure. For a given fiber volume fraction (or compressive strain value) σ_{eff} is given by the compressive stress-strain curve of the fiber network [1]. This curve is easily measured during a compression test on the dry fiber bed. When using prepregs, it is tedious and sometimes detrimental to the conservation of the original fiber lay-up to burn or dissolve the matrix to measure this curve. Methods are proposed in the literature to extract the compaction curve of the fiber bed from step-wise compression experiments of the prepregs, by letting the matrix bleed out at each step [2,3]. This curve can then be used to construct a process window for the prepreg compaction, knowing the initial fiber volume fraction or resin

content, and the level of applied pressure, using the approach proposed by Eom et al. [4,5], schematically described in Figure 1. Due to potential shrinkage of the resin during cure, for a given applied pressure during the process, any prepreg with fiber content higher than the intersect of the compaction curve with the pressure value would lead to a composite with voids, since there will be no pressure applied onto the resin during cure to compensate for the shrinkage. This intersection point would thus represent an optimal value, which could be reached either by using a resin content exactly matched to the process pressure, or by using a higher resin content and allowing limited bleeding. The fiber compaction curve thus represents the boundary between a sound composite (above the compaction curve) and one containing porosity (below the compaction curve).

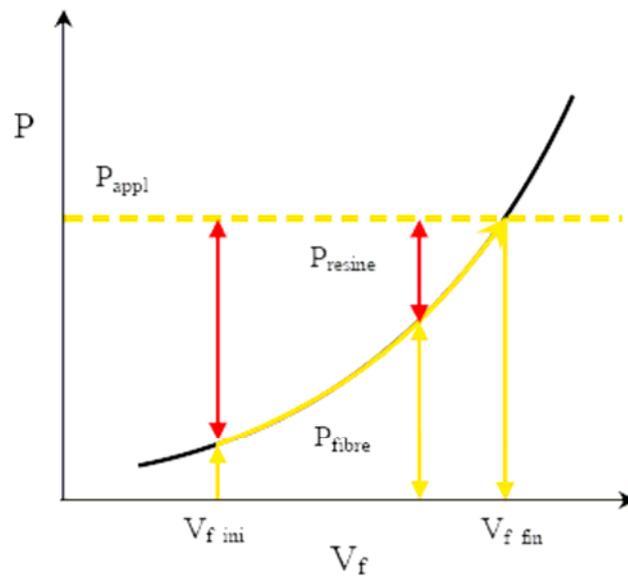


Fig. 1 Schematic view of the process window, the darker arrows represent the value of resin pressure for a given initial fiber volume fraction.

In addition, the influence of humidity in the prepreg could be added, following the methodology proposed by Kardos [6], which indicated, for a given relative humidity content RH in the epoxy based prepreg, that the pressure in the resin should be above $P_{\min} = 4.692 \cdot 10^3 \exp(-4892/T)$ (RH), where T is the cure temperature in K. This shifts the boundary line by a fixed amount above the fiber bed compaction curve, for a given value of humidity in the prepreg. The presence of volatils or entrapped air in the resin or between plies may as well be taken into account, if needed.

The present work thus proposes to construct and validate the process window for carbon-epoxy prepreps, with the goal to evaluate the maximal fiber volume fraction that could be reached for a given applied pressure to provide a sound composite. Several carbon-epoxy prepreg materials with fiber modulus ranging from 230 to 390 GPa were evaluated. Processing windows were established, based on the compaction curves of the fiber bed, for various fiber lay-ups. Composite parts were then manufactured with several resin contents, applied pressure, and humidity level to test the validity of the process map. As a consequence, limits in terms of fiber volume fraction can be set to the vacuum-bag production of carbon-epoxy composites, and potential routes to improve this limit will be discussed.

MATERIALS AND EXPERIMENTS

Materials

Three types of prepregs were used, as described in Table 1. The matrix was in all cases epoxy, prepreg A having a different matrix than B and C, and the fibers were carbon fiber of intermediate modulus.

Table 1 Description of the materials

Prepreg	Fiber modulus (GPa)	Fiber density	Resin content (wt%)	Areal weight (g/m ²)
A	390	1.82	26-32	300
B	230	1.8	34	300 or 200
C	242	1.81	34	160 or 200

Compaction experiments

The compaction experiments were performed following the method proposed in Ref.[2]. A compression fixture was designed, schematically presented in Figure 2, with a top piston applying load on the sample placed in the bottom part of the mould, which constrains the sample in the y direction, but not in the x direction, to allow flow of the resin out of the prepregs during the compression experiment.

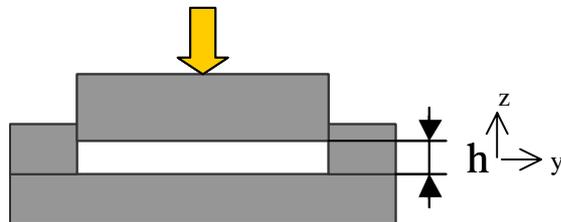


Fig. 2: Schematic of the compression fixture.

The fixture was mounted on a 100kN Interlaken hydraulic press, equipped with a convection oven to heat the assembly up to 100°C. The fixture temperature was monitored by two thermocouples. In a typical experiment, a stack of prepregs, either all aligned in the x direction, or laid at $\pm 45^\circ$ was placed in the fixture. The sample dimensions were 80 mm wide, 70 mm long, and about 6 mm high at the beginning of test, corresponding to about 16 to 30 layers depending on the fiber areal weight. The sample was then compressed in increasing steps of imposed displacement. The resulting load was recorded with the load cell. The displacement was recorded from the press position corrected for compliance effects. An example of curve is given in Figure 3. At each deformation increment, the load increased strongly, then decreased as the resin flowed out, to a stable position which corresponds to the equilibrium stress-strain curve of the fiber bed. The load curves were fitted as in Ref. [2] with an exponential decay. The number of steps, as well as the test temperature, were optimized for each resin type so as to get a best compromise between a low resin viscosity, and a long enough gel time.

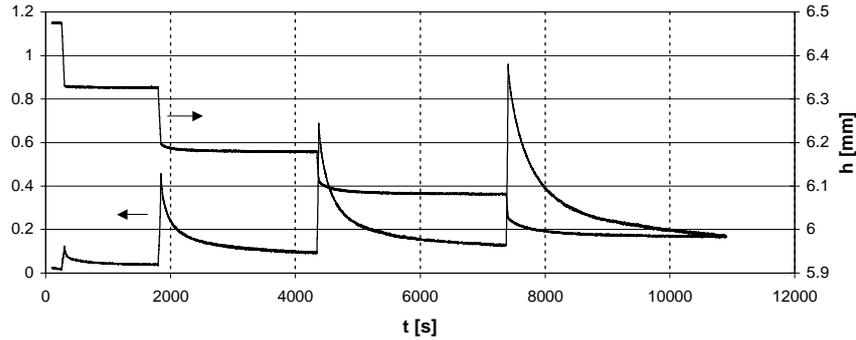


Fig. 3: Typical example of imposed displacement and measured load values with time.

Plate manufacturing

In parallel, several unidirectional plates, 1600 mmx1600 mm were manufactured in an autoclave, either under vacuum only, or using gas pressure up to 5 bars. The preparation of samples was as follows: an aluminum flat plate coated with demoulding agent was used as the base mould. The stack of preregs was laid on the mould, with intermediate vacuum compaction every three plies, up to about 18 plies. Consumables were then placed on top of the preregs. Different cases were tested, to investigate the effect of bleeding during cure: (1) a non-perforated film was placed directly on top of the prepreg, followed by a breather ply and vacuum bag, (2) a perforated film P3 (Aerovac) was placed directly on top of the preregs, followed by a breather ply and vacuum bag, (3) a non-impregnated peel-ply was placed directly on top of the prepreg, followed by a breather ply and vacuum bag. In all cases, the sides of the sample were sealed with sealant tape to ensure that all potential bleeding took place from the top of the preregs, to simulate the case of large parts. The samples were then cured in the autoclave, under vacuum and in some cases with additional pressure, following the recommended cure schedule for each prepreg type. The fiber volume content in the plates

was measured from the final average height of the plates, h , as follows: $V_f = \frac{n m_{of}}{\rho_f h}$, where n

is the number of plies in the laminate, m_{of} is the areal weight of each ply, and ρ_f is the fiber density. This was shown to be the most reliable way to compute this value, in order to compare it with the compaction measurements, which are also based on this calculation of V_f . The void content was evaluated using image analysis on optical microscopy photographs of polished cross-sections. To test the influence of humidity, a plate was prepared along the same route, using preregs which had been initially placed in a climatic chamber at 30° and 70%RH for one week.

RESULTS AND DISCUSSION

Compaction curves

Fig. 4 presents the compaction results for prepreg A, with a fiber orientation of 0° along the mould length, and of ±45°. The points were obtained in a series of several compression experiments. Some scatter is observed in the curves, which most probably results from several causes: there may be some variability between experiments in the fiber alignment, the width of the fiber assembly may slightly vary between experiments (although this is accounted for in the calculation of pressure and fiber volume fraction), and finally, it was shown that the exponential curve fitting used to calculate the

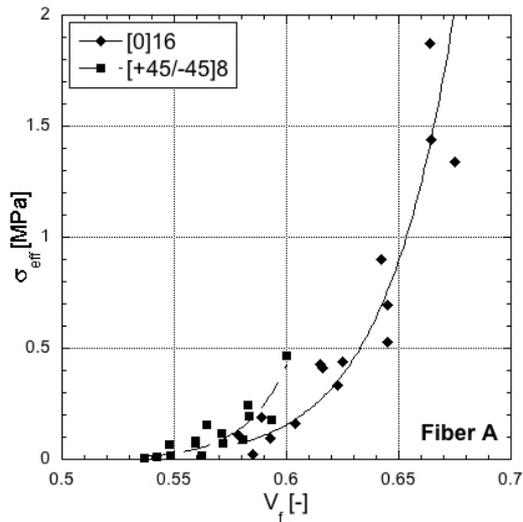


Fig. 4 Compaction results, prepreg A, 0° and ±45

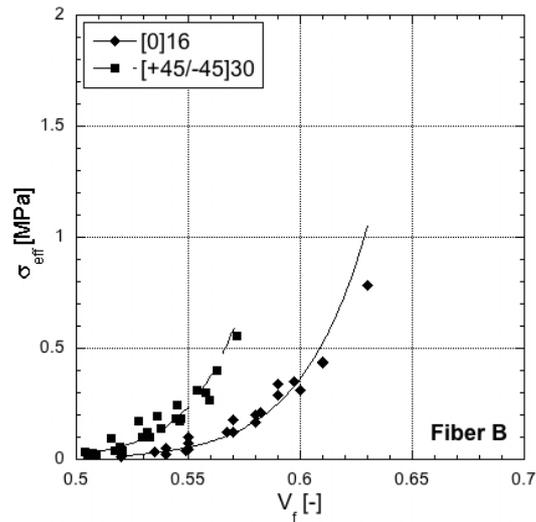


Fig. 5 Compaction results, prepreg B, 0° and ±45°

equilibrium points of the pressure curves sometimes tended to slightly overestimate the final pressure value. This error was estimated to 3%. However, a trend is clearly shown. The curves follow well the power law relation proposed by several authors [7-9], $\sigma_{\text{eff}} = k V_f^n$, where k is a constant which depends on the fiber bending rigidity. The value of the coefficient n is within the range observed for unidirectional fibers, 20 to 30, as shown in Table 2. As expected and already observed by Eom during compaction of dry glass fibers [5], the fiber lay-up with alternate orientations leads to a stiffer response of the fiber bed. This was however not observed in several other cases [2,3].

The following figures present the compaction results obtained for prepreg B (Fig. 5) and prepreg C (Fig. 6), at 0° and ±45°. Prepreg B follows the same trend as A, whereas C shows more scatter, and a less marked difference between the fiber lay-up type. This may be caused by the fact that these were made from heavy tow fibers, which initially leads to more misalignment in the layers. Finally, Fig.7 presents a comparison of the three prepreg types for the 0° direction. Whereas B and C follow almost exactly the same curve, A shows a better packing, that is a higher fiber volume fraction for a given applied pressure. This may be due to several causes: A fibers have a higher modulus, so they are likely to be more straight and less misaligned during prepreg manufacturing. Also, it is possible that more tension was applied on the fibers during prepreg manufacturing, leading to a denser packing.

Table 2 values of coefficients n for the compression experiments

Prepreg/layu	A 0	A ±45	B 0	B ±45	C 0	C ±45
p						
n	22	31	22	25	20	22

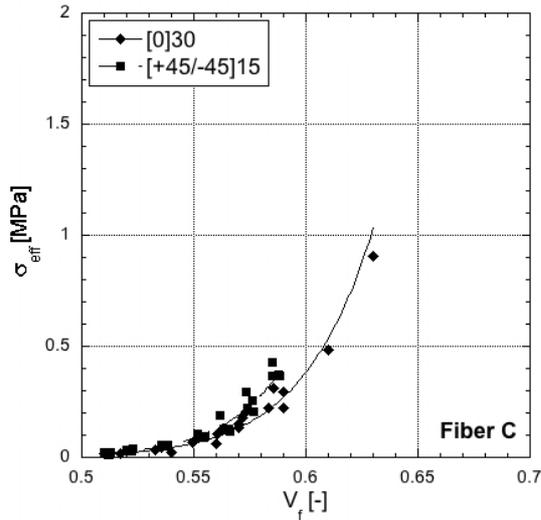


Fig. 6 Compaction results, prepreg C, 0° and ±45°

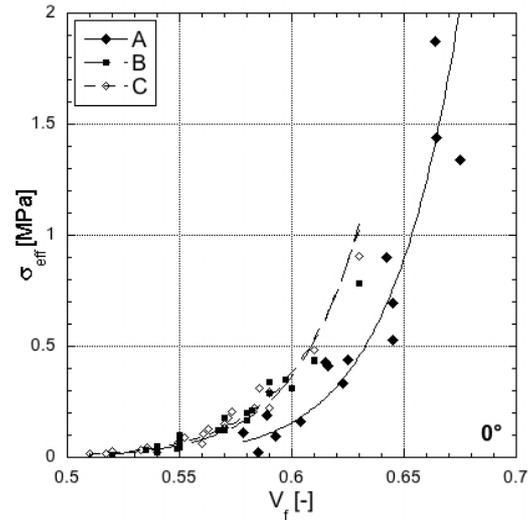


Fig. 7 Compaction results, comparison of all prepregs at 0°

Influence of pressure, initial resin content and consumables

Table 3 presents the various conditions of the plates manufactured, together with the measured final fiber volume fraction and void content. In most cases, the void content was very low, and it is reported as below 1%, to account for the measurement error. The vacuum bag processed samples lead to the highest void content. The samples processed after exposure to humidity in the same conditions as P9 are not reported below. They gave a very high void content, about $4.5 \pm 1.6\%$, indicating that humidity indeed exerts a strong influence on the part quality. However, a reference plate was made with the same prepregs, which were kept during one week in an air-conditioned room at 20°C before manufacturing the plate. This plate also had a final void content of $5.4 \pm 1.6\%$. As a consequence, it seems that the time out of the freezer of the prepreg exerts a much stronger influence than the humidity level. This point needs to be further clarified and taken into account when laminating large structures over several days.

For prepreg A, a study was conducted to test the influence of the release films and initial resin content on resin bleeding. As reported in Fig. 8, the samples with high resin content were subjected to bleeding when the consumables allowed it. The final fiber volume fraction thus varied from 56.8% when no bleeding was allowed, to 62.1% when a dry peel-ply was placed on the material. In the latter case, the final fiber volume fraction was close to that expected from the compaction curve for a pressure differential

Table 3 Conditions of manufactured plates

Sample number	P1	P2	P3	P4	P5	P6	P7	P8	P9	P10	P11
Prepreg type	A	A	A	A	A	A	A	A	B	C	C
Total applied pressure (bars)	5.9	5.9	5.9	5.9	5.9	5.9	2.9	2.9	0.9	0.9	3.9
Initial resin content (wt%)	32	32	32	26	26	26	32	26	34	34	34
Peel-ply type	1	2	3	1	2	3	1	1	2	2	2
Final V_f (%)	56.8	57.7	62.1	63.5	63.1	63.8	57.2	63.6	56	57	59.4
Void content (%)	<1	<1	<1	<1	<1	<1	<1	≈1	1.5	1.3	<1

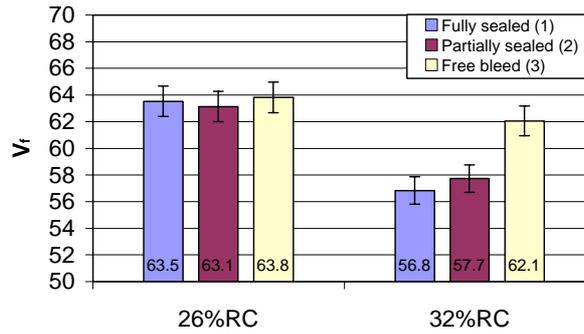


Fig. 8 Role of the consumables and initial resin content on the final fiber content in the plates.

of 5.9 bars, about 63%. The prepregs with the low resin content, calculated to achieve the maximal volume fraction at 5.9 bars (26 wt% resin should lead to $V_f=0.65$), all indeed had a final volume fraction fiber as expected, whether bleed was allowed or not.

PROCESS WINDOW

As a consequence, a process window can be constructed for each type of prepreg to estimate the risk of void content in the final part, for a given processing pressure, initial resin content and amount of allowed bleeding. An example is given in Fig. 9, for UD plates made from the prepreg A, reporting all samples, assuming that the humidity of the prepregs was negligible. It was verified that except sample P8, all plates had a very low void content. Several manufacturing strategies can thus be deduced, depending on the resin content of the prepregs and the amount of bleeding allowed, based on the consumable type and the resin viscosity profile, to reach the maximal fiber volume fraction for a given pressure differential. Fig.10 reports the process window for prepreg B and C, assuming no humidity, or 70%RH for a cure at 100°C, using Ref.[6]. The points corresponding to plates performed under vacuum only fall very close to the pressure boundary, close as well to their theoretical fiber volume fraction $V_f=0.56$ based on their initial resin content, and below the boundary with 70%RH. In that region, the curve is flat, and the role of volatils and humidity cannot be ignored anymore. In addition, the exact value of the pressure differential is more difficult to assess, in case of a leak in the mold or the vacuum bag, for example. The process window is however of use to assess the maximal volume fraction that can be achieved with a given prepreg under vacuum bagging, and the risks of a given humidity content.

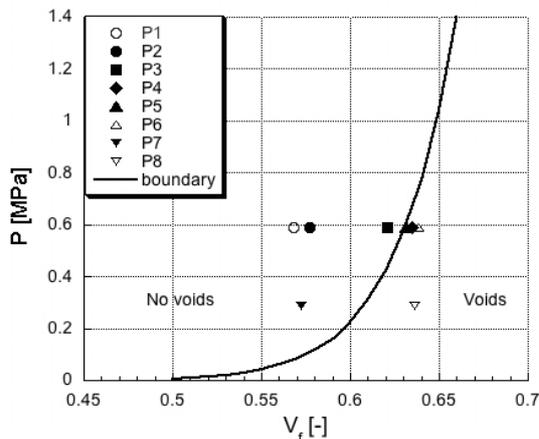


Fig.9 Process window for prepreg A

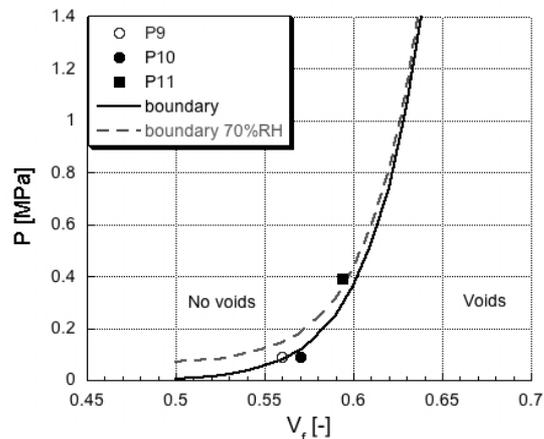


Fig.10 Process window for prepreg B/C

CONCLUSION

The present work showed that process windows for sound processing of carbon-epoxy composites can be established, simply based on compaction curves of the initial prepreg. It is worthy to note that this curve could also be established from the final volume fraction of plates cured with free resin bleed at various applied pressures, if a heated press is not available. Several strategies can be devised based on these guidelines to process the material, depending on the initial resin content and applied pressure during processing. The use of peel-plies and other consumables then allows the amount of resin bleed to be adjusted to reach the maximal fiber fraction. Comparison of prepregs showed that significant differences can be observed, for a given direction, depending on the fiber type and possibly on the prepreg manufacturing route. As a consequence, it should be possible to reach a fiber volume fraction up to 60% with vacuum-bag processing if the fibers are initially well packed during prepreg manufacturing. This does not hold, however, if the fiber orientation is alternated during the lay-up, as is usually done when manufacturing real parts. Again, the choice of prepregs, such as A or C for example rather than B may help achieve higher fiber content and a sound composite.

ACKNOWLEDGEMENTS

This work was funded in part by the Swiss Commission for Technology and Innovation contract n°6972.2, and in part by the Swiss National Fund for Scientific Research contract n°200020-105169. Alinghi SA and Décision SA are thanked for donating prepregs and for fruitful discussions.

REFERENCES

1. V. Michaud and A. Mortensen, "Infiltration processing of fiber reinforced composites: governing phenomena", *Composites part A*, 32, pp.981-996, 2001.
2. P. Hubert, A.Poursartip, A method for the direct measurement of the fibre bed compaction curve of composite prepregs", *Composites Part A*, Volume 32, , Pages 179-187 (2001).
3. T. G. Gutowski, *Advanced composite manufacturing*, John Wiley & Sons Inc. (1997)
4. Y. Eom, L. Boogh, V. Michaud, P. Sunderland and J.-A. E. Månson. "Stress-initiated void formation during cure of three-dimensionally constrain thermoset resin". *Polymer Engineering and Science*, Vol.41, No.3 (2001).
5. Y.Eom, L.Boogh, V.Michaud and J.A-E. Månson, "A structure and property based process window for void-free thermoset composites", *Polymer Composites*, vol.22, N°1, Pages 22-31(2001).
6. J.L. Kardos, M.P. Dudukovic and R. Davé, "Void growth and resin transport during processing of thermosetting matrix composites", *Advances in Polymer Science*, Vol.80, K. Dusek, ed, Springer Verlag, Berlin, (1986).
7. Toll, S. and Månson, J.-A.E., "Elastic compression of a fiber network", *J. Appl. Mech.*, Vol. 62, 1995, pp. 223-226.
8. van Wyk, C.M., "Note on the compressibility of wool", *J. Textile Institute*, Vol. 37, 1946, pp. T285-T292.
9. C. Servais, V. Michaud and J.A-E. Månson, "The packing stress of impregnated fiber mats", *Polymer Composites*, Vol.22, n°2, April 2001, pp.298-311.