FATIGUE PROPERTIES OF HIGHLY ORIENTED POLYPROPYLENE TAPES AND ALL-POLYPROPYLENE COMPOSITES

N.-M. Barkoula[1,2], B. Alcock[1,3], N. O. Cabrera[1,3], and T. Peijs[1,3]

[1] Queen Mary, University of London, Department of Materials, Mile End Road E1 4NS, London, UK
[2] Department of Materials Science and Eng. University of Ioannina, GR-45 110, Ioannina, Greece
[3] Eindhoven University of Technology, Eindhoven Polymer Laboratories, PO Box 513, 5600 MB Eindhoven, The Netherlands

ABSTRACT

This paper describes the fatigue behaviour of newly developed all-polypropylene (all-PP) tapes and composites, with reference to the composite processing conditions and compares this behaviour with commercial alternatives. All-PP tapes are highly oriented and their failure behaviour follows that of other highly oriented polymers. All-PP woven composites fail ultimately due to PP tape failure. However, this failure mode is preluded by delamination of fabrics in the woven structure. Consolidation pressure plays a decisive role in controlling the interlaminar properties and hence controls the delamination resistance and furthermore the fatigue limit of the composite. Comparison of all-PP woven composites with commercial alternatives based on glass and natural fibres reveals the excellent relative performance of all-PP composites under fatigue loads. Fatigue properties of all-PP composites are however sensitive to the testing temperature and elevated temperatures can lead to a rapid reduction of the fatigue resistance of these all-polymer systems.

1. INTRODUCTION

Quite recently, all-polypropylene (all-PP) or self-reinforced composites have been proposed to replace traditional fibre reinforced PP composites (glass fibre (GF)/PP or natural fibre (NF)/PP) for automotive applications due to their environmental friendly nature. All-PP composites have specific economic and ecological advantages over composites based on GF or NF since, upon recycling of all-PP composites, a polypropylene blend is obtained which can be reused to make again all-PP composites, or alternatively, be used for other PP-based applications. The issue of recycling is of particular relevance to the automotive industry with the recent End-of-Life Vehicle Directive of the European Union aiming to increase the rate of recycling of all vehicles to at least 95% by average weight by 2015 [1].

Although recyclability is a very important issue, cost of manufacturing and mechanical performance remain of key importance for many engineering applications, including the automotive industry. One of the possible routes for manufacturing of single polymer composites was proposed by Peijs and co-workers [3-4] based on the consolidation of co-extruded fibres/tapes. The newly developed all-PP composites make use of high modulus PP homopolymer tapes, which are co-extruded with a thin coating of a PP copolymer. This means that all-PP composites possess a high volume fraction (~90%) of highly oriented PP reinforcement fraction and ~10% volume fraction of copolymer layer which acts as matrix. A temperature-processing window of >30°C can be achieved using this method. The short-term mechanical properties of such all-PP composites (unidirectional (UD) and woven) were recently studied in detail [3-4]. These studies reveal the feasibility of all-PP composites as replacement of traditional PP reinforced composites, under static loads. In this study the long-term performance of these all-polymer composite systems under cyclic loads (i.e. fatigue) is presented.

Fatigue of materials has enormous practical implications since in real applications service loads are rarely purely static. It has been estimated that over 80% of all service failures of structural materials can be traced to fatigue [5]. Failure development due to fatigue of oriented polymers (i.e. fibres) and polymer composites is a rather complex phenomenon, which mainly depends on the loading conditions, as well as the nature of the examined systems. In case of oriented fibres, the degree of macromolecular orientation, the level of crystallinity, the structure and morphology of the sample, as well as the testing temperature are decisive parameters for their fatigue life [6-10]. For polymer composites, the performance of the fibre-matrix adhesion [11-18], the fibre properties, fibre volume fraction and fibre type [18-20], the fibre distribution and lay-up configuration [14, 20-22] and the matrix properties [14, 23, 24] are the most influencing parameters. Nevertheless, it should be noted, that not only internal parameters (material properties) but also external ones, such as manufacturing/processing conditions and service temperature [25, 26], could have a great influence on the fatigue life of a composite system.

Since fatigue performance is of key importance for the design in industry, the main scope of this study is to investigate the fatigue behaviour of all-PP tapes and woven composites, to recognise and understand the failure mechanisms that prevail during fatigue. As these composites are to replace conventional composites such as glass mat reinforced thermoplastics (GMT) and natural fibre mat reinforced thermoplastics (NMT) a comparison is made with the fatigue behaviour of these materials. It is important to note that in both GMT and NMT composites reinforcement is randomly oriented, whereas in all-PP composites it is bidirectionally oriented.

2. EXPERIMENTS

2.1. Material Manufacturing

The tapes throughout this investigation are co-extruded three layer tapes, with a A:B:A (copolymer:homopolymer:copolymer) structure, and are manufactured by Lankhorst Indutech BV (The Netherlands). The tape properties are summarised in Table 1.

Table 1. *Summary of PP tape properties*

Width	2.15 mm
Thickness	60 μm
Density, ρ	0.732 $g.cm^{-3}$
Draw Ratio, λ	17
Composition [A:B:A]	5.5:90:5.5

These tapes have been woven into a plain weave fabric with a density of ~100 $g.m^{-2}$ by BW Industrial BV (The Netherlands). Previous investigations showed that manufacturing conditions (consolidation pressure) play a very important role on the static properties of all-PP woven composites [3]. To establish the effect of varying compaction pressure on the properties of all-PP composites, two different techniques were used to consolidate the woven fabrics into composite plates. For high pressures (around 2.5 MPa), a belt press technology was used (IVW Kaiserslautern, Germany). A compaction temperature of 145 ºC was found appropriate to get a good consolidation. The specimens will be referred hereafter as belt-pressed woven all-PP composites (BP-all-PP). To accurately compact specimens at low pressures (around 0.1 MPa), the vacuum bagging technique was employed. Compaction temperature is again 145 ºC. These specimens will be referred as vacuum-bagged woven all-PP composites (VB-all-PP). Previous references give details on both manufacturing techniques [6]. In addition to PP tapes and all-PP composites, two types of fibre reinforced PP are used for comparison. The first is Symalit®, (hereafter referred to as GMT) and is a random glass fibre reinforced PP with a fibre weight fraction of 23% while the second is a random natural (flax) fibre reinforced PP (hereafter referred to as NMT) with a fibre weight fraction of 30%, both manufactured by Quadrant Composites, Switzerland.

2.2. Testing procedure

The static tests on highly oriented PP tapes and all-PP composites were performed on an Instron 5584 tensile testing machine. The machine was equipped with different load cells (i.e. 500N for the single tape tests and 10kN for the composite specimen test) and a data acquisition computer. All tensile tests were performed at 5mm.min^{-1}. Each test was performed at least three times. All-PP tapes had a length of 250 mm. The dimensions of the test coupons are 15 mm x 210 mm x 1.7 mm for all-PP woven, while GMT and NMT test specimens where of similar dimensions but of different thickness, 2 and 4 mm respectively.

Fatigue tests were performed on an Instron 8500 servo-hydraulic test machine. The system was operating under load control, applying a harmonic tensile stress with constant amplitude. Tension-tension fatigue tests were carried out throughout this study at a frequency of 3Hz and a stress ratio R of 0.1. In order to avoid heating effects a frequency of 3Hz was selected. A thermocouple was occasionally applied on the specimen surface during testing, and the increase in temperature was monitored. The temperature increase never exceeded 3°C for all specimens. Global isothermal testing conditions can therefore be assumed. Fatigue tests were carried out at different stress levels between the ultimate tensile strength (UTS) and the fatigue limit, resulting in so-called Wöhler or S-N curves, which show the relation between the maximum stress and the number of cycles to failure. Tests exceeding the 10^6 cycles were terminated. Specimens that failed in or closed to the grips were discarded. The geometry of the all-PP samples was the same as those used for the tensile characterisation.

3. EXPERIMENTAL RESULTS & DISCUSSION

3.1. Fatigue response of PP tapes and all-PP composites

The fatigue behaviour of materials is conventionally characterised by a Wöhler or S-N curve. It has been suggested that the overall S-N curve of both highly oriented fibres and fibre reinforced polymer composites has three stages, the initial stage (Stage A), the progressive stage (Stage B) and the final stage or fatigue limit (Stage C). The fatigue response of PP tapes (S-N curve) is shown in Figure 1a. In addition, a micrograph of the macroscopic failure of the tapes after fatigue is presented in Figure 1b. As mentioned in paragraph 2.1, PP tapes consist of 90 wt.% oriented phase, therefore they are considered as highly oriented polymer fibres. Highly drawn PP tapes possess a highly fibrillar character which is reflected also on their fatigue response. Figure 1 confirms this consideration, since it shows a typical S-N pattern common to highly oriented polymers. In this pattern, the initial stage, in which considerable structural change takes place at a relative high rate and thus the curve decreases at a relatively low rate, is absent. However, Stages A and B are not always separable. Stage B, in which the curve shows an approximately linear decrease with the logarithm of N appears from the beginning of the fatigue test. This is mainly due to the fact that PP tapes tested here are highly oriented and there is not so much capacity for further orientation and structural changes during Stage A of fatigue, which usually occur in the amorphous phase of the fibres [10]. Thus, fatigue rupture most likely occurs due to repeated sliding motions of the fibrillar elements, which damage the interfibrillar bonding but fatigue rupture can also be due to rupture of tie chains in the amorphous regions of the macromolecules [10]. The fatigue limit of PP tapes is observed to be at stresses as high as 400MPa, which represents around 65 % of the UTS of the material and is quite high compared to values obtained in case of other highly oriented polymer fibres [6-10].

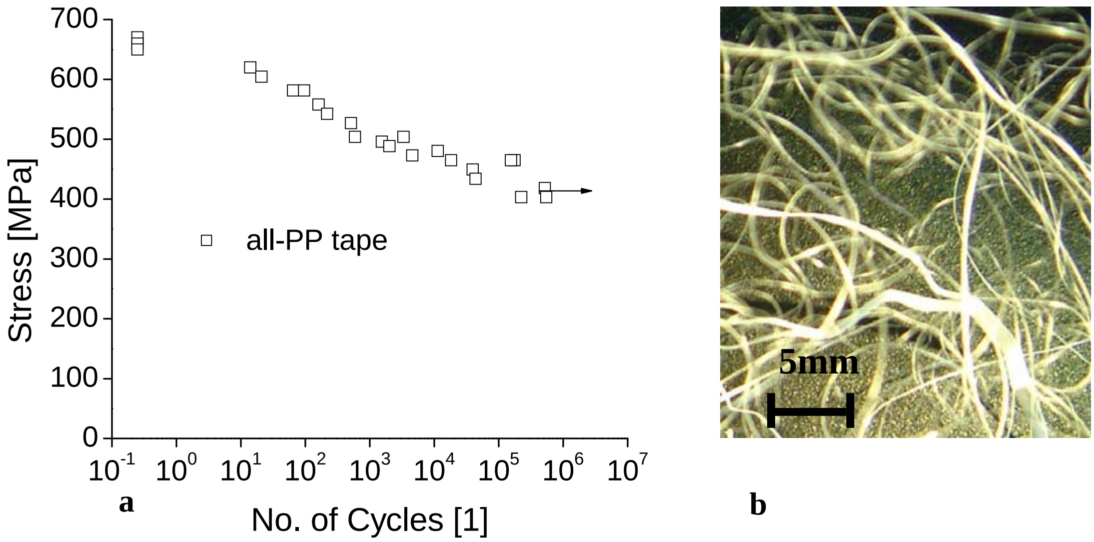

Figure 1. (a) Fatigue response of PP tapes at RT represented by S-N curves (arrows signify specimens that did not fail after 10^6 cycles) and (b) macroscopic image of a failed PP tape after failure, showing the completely fibrillated tape structure and no recognisable tape geometry remaining.

Furthermore, the fatigue properties of all-PP composites are tested on the example of belt-pressed (BP) and vacuum bagged (VB) all-PP specimens. Figure 2 demonstrates the fatigue response of these materials as a function of the number of cycles to failure. The first observation that can be made is that both specimens behave similarly under fatigue with the only difference at a first approximation that the curve of the VB-all-PP specimen is shifted at lower stress levels. It is therefore confirmed that consolidation pressure has a great influence on the fatigue response of all-PP woven composites. This result was expected, since, it is already known that consolidation pressure, especially when reduced to as low as 1 bar, has a very strong effect on the static properties and failure process during static loadings [3]. Acoustic emission (AE) results [4] show that there is a large effect of consolidation pressure on the initiation of damage through delaminations, which are more extensive and occur earlier in case of VB-all-PP composites.

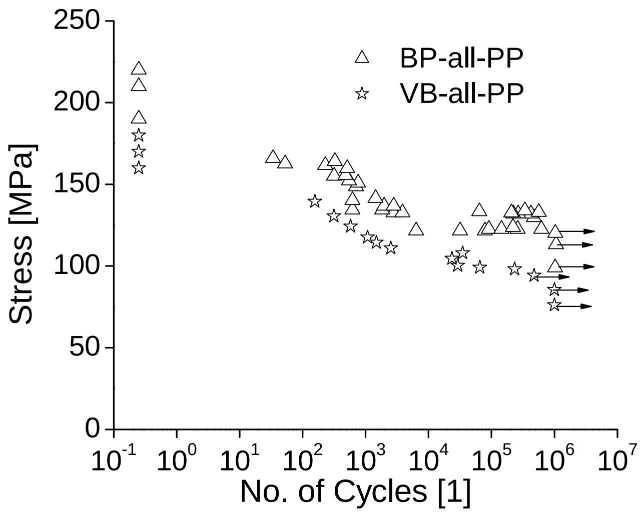

Figure 2. S-N curves for BP-all-PP, and VB-all-PP woven composites, tested at RT, illustrating the effect of consolidation pressure on the fatigue response. Arrows signify specimens that did not fail after 10^6 cycles.

This is the reason of the shift of the fatigue curve to lower values in case of VB-all-PP composites. It is interesting to note, however, that the consolidation pressure does not seem to alter the ultimate failure mechanisms during fatigue, since both BP-all-PP and VB-all-PP specimens have the same course to failure. This observation will be discussed further in a separate paragraph of this paper dedicated on the damage development in all-PP composites. The second interesting result is that the random fibre breakage mechanism of Stage A, usually observed in polymer composites, is totally absent and the progressive fatigue damage mechanism (Stage B) is immediately operative. Two main reasons are responsible for this. The first one is clearly related to the very narrow strength distribution of all-PP tapes as discussed earlier and Stage A, which is associated with the scatter band in the fibre properties diminishes. Next to that, previous observations on the example of GF/PP composites have shown that when the matrix of the composite system is ductile, the first region of fatigue tends to disappear anyway because the fibre bridging crack-arresting mechanism are not active or at least less effective [14, 15]. For both BP-all-PP and VB-all-PP specimens, the endurance limit is found to be at about 64% of the UTS of each specimen. It differs however in its absolute value, which in case of the BP material is at around 125MPa while in case of VB-all-PP material at 90 MPa.

For a direct comparison of the fatigue behaviour of PP tapes with all-PP composites (BP- and VB-all-PP), a normalised "S-N" curve was created and their behaviour was presented in a single graph. The stress was normalised over the UTS of each material and plotted against the number of cycles to failure. From this plot (Figure 3), speculations about the underlying mechanisms leading to fatigue can be made. The fact that the curve of the tape almost coincides with those of the BP-all-PP and VB-all-PP specimens suggests a similar strength reduction rate for all specimens, which indicates that similar fatigue mechanisms prevail. This means that there is no tape governed crack growth through subsequent layers, and the fatigue life of the materials depends mainly on the fatigue characteristics of the tape. As already seen from results on the fatigue response of consolidated composites, and stated in previous part of this work, interface/delamination damage occurs, however this damage does not seem to be critical for the ultimate failure of the composites. It is furthermore suggested that failure occurs when the strain to failure of the tapes is exceeded.

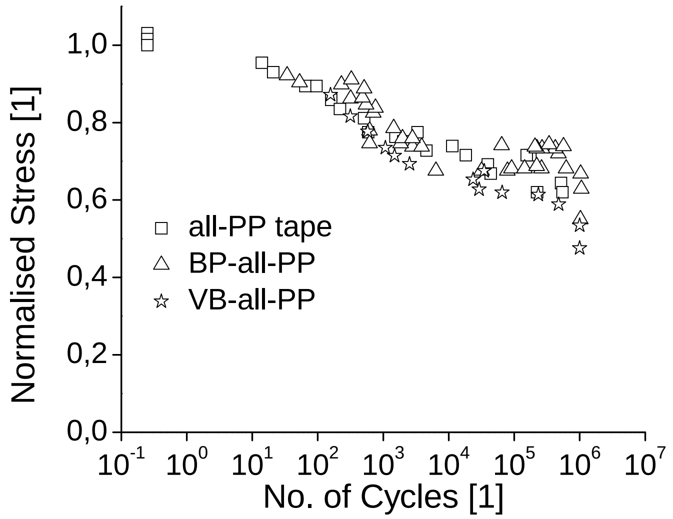

Figure 3. Normalised stress (σ/σ_o) as a function of the number of cycles to failure, of PP-tapes, BP-, VB-all-PP woven composites comparing the fatigue response and the damage development of these materials. σ_o corresponds to the UTS of respective material.

3.2. Comparison with GMT and NMT

Previous studies showed that the mechanical properties of all-PP woven composites under static load, compared well to those of other commercial PP composites such as glass mat reinforced PP (GMT), and a flax fibre mat reinforced PP (NMT) [6]. In this section, a comparison of these materials is made under fatigue loads. Clearly, one of the advantages of all-PP composites over GMT and NMT materials is the much higher reinforcement component (appr.90% vs. 30%). Next to that, as mentioned before, reinforcement is bidirectional rather than random in all-PP composites. The exceptionally high reinforcement content that originates from the tape structure is one of the keys to the success of these materials as on a one-to-one basis PP tapes can never compete with glass fibres in terms of strength and stiffness. However, in the final composite system this lower 'fibre' performance is more then compensated by the much higher fibre volume fraction which is not achievable in traditional composites. The normalised S-N curves were plotted for the fatigue properties of BP-all-PP systems, GMT and NMT and can be found in Figure 4. From this figure, it is obvious that all-PP composites possess superior fatigue resistance, while GMT presents the highest reduction rate. The damage process during deformation of all-PP composites seems to be very much different compared to that of glass fibre reinforced PP.

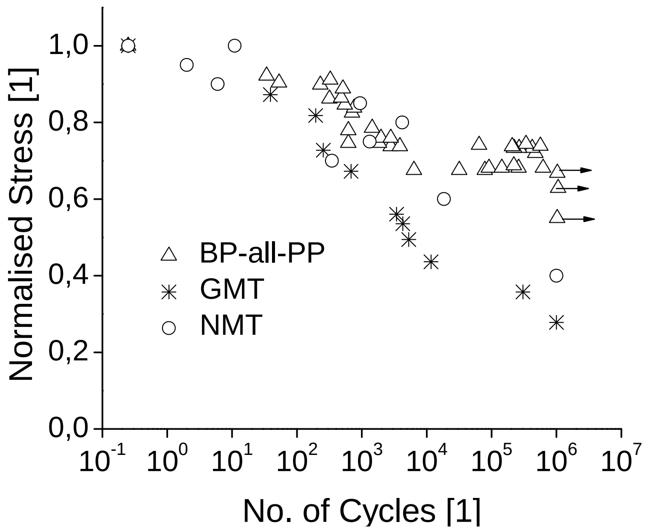

Figure 4. Normalised stress (σ/σ_o) as a function of the number of cycles to failure, comparing the fatigue response and the damage development of BP-all-PP woven composites with GMT and NMT. σ_o corresponds to the UTS of respective material. Arrows signify specimens that did not fail after 10^6 cycles

4. CONCLUSIONS

The tension-tension fatigue properties of PP tapes and all-PP woven composites have been studied here and the possible damage development mechanisms have been discussed. Both PP tapes and consolidated all-PP woven composites have quite high resistance to fatigue and their endurance limit is as high as 65% of their UTS. High resistance to fatigue of all-PP composites is linked to the low variability in strength of PP tapes, and good interfacial properties. In all-PP woven composites, failure occurs always through tape breakage facilitated by delaminations. However, the level of consolidation is crucial for the extent of delaminations and for the achieved fatigue limit.

The fatigue behaviour of All-PP composites was compared with commercial GMT and NMT composites, where the endurance limit varied between 30% and 40% of their UTS, respectively. This comparison reveals that all-PP woven composites show better strength retention after fatigue and this makes them feasible for applications where tension-tension fatigue is operative. Furthermore PP tapes due to their fibrillated structure and their ductility, do not act as stress concentration points, where cracks initiate and propagate, which is the case in fatigue of glass fibre reinforced composites.

ACKNOWLEDGEMENTS

The co-extruded PP tapes used in this study were kindly supplied by Lankhorst Indutech BV, Netherlands, and woven into fabrics by BW Industrial BV, Netherlands. This work is partly sponsored by the Dutch Government's Economy, Ecology and Technology (EET) programme for sustainable development, under grant number EETK97104. One of the authors (N.-M. B.) would like to acknowledge the EU (ORTHOFLEX Growth project, no: G5ST-CT-2002-50185) for financial support.

REFERENCES

1. Directive 2000/53/EC of European Parliament of the Council of 18 September 2000 on End-of-Life Vehicles. 2000

2. Cabrera, N, Alcock, B, Loos, J, Peijs, T, "Processing of All-Polypropylene Composites for Ultimate Recyclability", Proceedings of the Institute of Mechanical Engineers: Journal of Materials: Design and Applications, 2004; 218: 145-156.

3. Alcock, B, Cabrera, NO, Barkoula, N-M, Loos, J, Peijs, T, "The Mechanical Properties of Unidirectional All-Polypropylene Composites", Composites: Part A 37 (5), 2006; 716-726.

4. Alcock, B, Cabrera, NO, Barkoula, N-M, Spoelstra, AB, Loos, J, Peijs T, "The Mechanical Properties of Woven Tape All-Polypropylene Composites", Composites: Part A, 2007; 38 (1): 147-161.

5. Dauskardt, RH, Ritchie, RO, Cox, BN, "Fatigue of Advanced Materials: Part 1", Advanced Materials and Processes, 1993; 26-31.

6. Kaiya, N, Takahara, A, Kajiyama, T, "Fatigue Fracture Behaviour of Solid-State Extruded High-Density Polyethylene", Polymer Journal 1989; 21: 523-531

7. Kaiya N, Takahara A, Kajiyama T. Fatigue Fracture Behaviour of Oriented Ultra-High Molecular Weight Polyethylene. Polymer Journal 1990;22: 859-865

8. Jo N-J, Takahara A, Kajiyama T. Analysis of Fatigue Behaviour of High-Density Polyethylene based on Non-Linear Viscoelastic Measurements Under Cyclic Fatigue. Polymer Journal 1993;25: 721-729

9. Jo N-J, Takahara A, Kajiyama T. Effect of Crystalline Relaxation on Fatigue Behaviour of the Oriented High-Density Polyethylene Based on Non-Linear Viscoelastic Measurements. Polymer Journal 1994; 26: 1027-1036

10. Narisawa I, Ishikawa M, Ogawa H. Fatigue Processes in Highly Oriented Nylon-6 Fibers. Journal of Polymer Science 1997;15: 1055-1066

11. Owen MJ. Fatigue Processes in Fiber Reinforced Plastics. Philosophical Transactions of the Royal Society 1980;A294: 535-543

12. Gassan J, Bledzki AK. The Influence of Fiber-Surface Treatment on the Mechanical Properties of Jute-Polypropylene Composites. Composites: Part A 1997;28: 1001-1005

13. Youssef Y, Denault J. Thermoformed Glass Fibre Reinforced Polypropylene Microstructure, Mechanical Properties and Residual Stresses. Polymer Composites 1998;19: 301-309

14. van der Oever M, Peijs T. Continuous-Glass-Reinforced Polypropylene Composites II. Influence of Maleic-Anhydride Modified Polypropylene on Fatigue Behaviour. Composites: Part A 1998;29: 227-239

15. Gamstedt EK, Berglund L, Peijs T. Fatigue Mechanisms in Unidirectional Glass-Fibre-Reinforced Polypropylene. Composites Science and Technology 1999;59: 759-768

16. Gassan J, Bledzki AK. Possibilities to Improve the Properties of Natural Fibre Reinforced Plastics by Fibre Modification (Jute Polypropylene Composites). Applied Composite Materials 2000;7: 373-385

17. Gassan J. A Study of Fibre and Interface Parameters Affecting the Fatigue Behaviour of Natural Fibre Reinforced Composites. Composites: Part A 2022;33: 369-374

18. Wambua P, Ivens J, Verpoest I. Natural Fibres: Can they Replace Glass in Fibre Reinforced Plastics? Composites Science and Technology 2003;63: 1259-1264

19. Talreja R. Fatigue of Composite Materials: p3-58. Tecnomic Publishing Co., Lancaster, PA 1987

20. Gassan J. A Study of Fibre and Interface Parameters Affecting the Fatigue Behaviour of Natural Fibre Reinforced Composites. Composites: Part A 2002;33: 369-374

21. Ferreira JA, Costa JDM, Reis PNB. Static and Fatigue Behaviour of Glass-Reinforced Polypropylene Composites. Theoretical and Applied Fracture Mechanics 1999;31: 67-74

22. Ferreira JA, Costa JDM, Reis PNB, Richardson MOW. Analysis of Fatigue and Damage in Glass-Fibre-Reinforced Polypropylene Composite Materials. Composites Science and Technology 1999;59: 1461-1467

23. Talreja R. In: Structure and Properties of Composites, Vol 13, ed. T W Chou: p383. VCH, Weinhein 1993

24. Bureau MN, Denault J. Fatigue Behaviour of Continuous Glass Fiber Composites: Effect of the Matrix Nature. Polymer Composites 2000;21: 636-644

25. Bureau MN, Denault J. Fatigue Resistance of Continuous Glass Fiber/Polypropylene Composites. Composites Science and Technology 2004;64: 1785-1794

26. Bureau MN, Perrin AR, Denault J, Dickson JI. Intralaminar Fatigue Crack Propagation in Continuous Glass Fiber/Polypropylene Composites. International Journal of Fatigue 2002;24: 99-108