ELSEVIER

Composites: Part A 33 (2002) 453–458

composites
Part A: applied science and manufacturing

www.elsevier.com/locate/compositesa

Fatigue characterization of polyethylene fiber reinforced polyolefin biomedical composites

M. Kazanci[a], D. Cohn[a], G. Marom[a,*], C. Migliaresi[b], A. Pegoretti[b]

[a]*Casali Institute of Applied Chemistry, The Hebrew University of Jerusalem, Givat Ram Campus, 91904 Jerusalem, Israel*
[b]*Department of Materials Engineering, The University of Trento, 38050 Mesiano, Trento, Italy*

Received 23 April 2001; revised 30 October 2001; accepted 12 December 2001

Abstract

Filament wound flat strip composites of polyethylene fiber reinforced ethylene–butene copolymers were produced and their fatigue behavior under cyclic loading was studied. Three different copolymer compositions and two different winding angles were employed in order to study the effects of branching density in the polymeric matrix and of reinforcement angle on the fatigue response of the composite. The results were in agreement with published fatigue models, showing that the short-term fatigue behavior, at relatively high stress levels, was controlled by the static properties of the materials, exhibiting better fatigue resistance for lower branching density of the copolymer and for a smaller reinforcement angle. However, the long-term fatigue behavior, at moderate stress levels, was governed by the fatigue rate of degradation, which decreased with the branching density and winding angle. The fatigue induced creep resulted in fiber reorientation in the loading direction, which in turn generated high residual properties. It was concluded that various polymer/angle combinations could result in fatigue-proof composites of significant residual properties at 10^6 fatigue cycles. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: A. Polymer-matrix composites (PMCs); B. Fatigue; E. Filament winding; Polyethylene–butene copolymers

1. Introduction

The fatigue behavior of advanced composite materials has been studied extensively to address problems resulting from their exposure to long-term engineering service conditions. Mechanical fatigue is the most common type of failure of composite structures in service [1], where the polymeric matrix develops brittle cracks, which are usually generated by low stresses applied over a long time period [2,3]. In general, two fatigue failure modes are recognized depending on the stress angle relative to the fiber direction. The first, for 0°, is fiber dominated and the second, for angles above 0°, is interface and matrix dominated. For angle ply composites, a combination of factors such as the static strength, the strength of the interface and the viscoelastic nature of the matrix, determines the fatigue performance. Obviously, due to much higher properties of the fibers, higher fatigue stresses are endured for smaller angles, for which the static strength of the composite is governed by the properties of the fibers and their volume fraction [4]. However, the overall fatigue resistance is determined by the rate of degradation (defined by the slope of the S–N plot, where S is the stress and N is the number of cycles to failure), which is matrix and interface dependent. As a result, the fatigue behavior of the angle ply composites is controlled mostly by the properties of the matrix and of the fiber-matrix interface. In general, the rate of strength degradation has been taken to be inversely proportional to some powers of the residual strength and, on this basis, a relationship between the initial strength and the residual strength has been derived [5]. The fatigue limit is defined as the stress corresponding to the boundary between the propagating and the non-propagating matrix cracks at 10^6 cycles. The fatigue ratio is expressed by the ratio of the fatigue limit to the static fracture stress. This ratio is a useful index for rating fatigue properties of composite materials [5].

Thermoplastic matrices, such as polyethylene (PE), could offer advantages over thermosets. Whereas, damage in the latter occurs by propagation of small localized fiber-bridged cracks, the first show more extensive and distributed damage, plastic flow and fiber reorientation with progressive fiber breakage [6]. Moreover, the fracture toughness of the interface between the fiber and the matrix could be improved by giving it more ductility. In addition to PE, the scientific literature reveals considerable interest in short-chain branched PE, because of its outstanding fracture

---

* Corresponding author. Tel.: +972-2-658-5898; fax: +972-2-658-6068.
E-mail address: gadm@vms.huji.ac.il (G. Marom).

PII: S1359-835X(02)00002-7

Table 1
Characteristics of the PE–butene copolymer matrices

Matrix	Branching[a] (per 1000 main chain C atoms)	T_m (°C)	Density (g/cm^3)	Modulus (MPa)	Crystallinity (%)
Exact 4041	66	59.7	0.878	22	10
Exact 4011	50	70.0	0.888	30	13
Exact 4015	42	82.6	0.894	42	17

[a] Calculated from the Exact technical information.

and mechanical properties, such as failure time under static fatigue and environmental stress crack resistance. For example, the failure time under static fatigue of an ethylene–hexene copolymer in the air is approximately 100–1000 times longer than that of low-density polyethylene (LDPE) and high-density polyethylene (HDPE). This dramatic improvement in static fatigue has been attributed to the presence of short-chain branches in the molecules of PE. Thus, an increase in the branch concentration also causes more tie molecules to be formed which results in higher resistance of the molecules against pulling through the crystalline region. Accordingly, the dynamic fatigue of several ethylene–hexene copolymers was found to increase by approximately four orders of magnitude when the branch concentration increased from 0 to 4, 6 butyl branches per 1000 carbons [3,7]. However, it has also been proved that a negative correlation between fatigue and constant stress lifetimes may prevail in such copolymers when the branches form clusters, and that if instead the branches are more uniformly spaced, they will be more effective in preventing fracture [8]. These facts are compatible with the observations that the fracture toughness of the copolymers can be increased by increasing the level of short-chain branching and the length of side chains and by decreasing the crystal thickness [3,7–11].

The question of fatigue performance of engineering composites becomes critical when they are destined for biomedical applications as orthopedic prostheses [12]. In particular, fatigue and wear damage of total joint components has been recognized as significant clinical problems limiting the lifetime of joint arthroplasties [13]. In this context, various studies exist on the long-term performance of HDPE, and UHMWPE fibers, designated for tendons, ligaments and joint prostheses [12–14].

The objective of this work is to present and discuss a new type of filament wound, flat strip composites of PE fiber reinforced polyolefins for biomedical applications, namely, for ligament and tendon implants. It follows two preliminary studies by these research groups of the fatigue behavior [15] and relaxation processes [16] of similar PE/PE composites. Here, it is intended to demonstrate the versatility of the new product by involving different polyolefin compositions based on ethylene–butene copolymers and various winding angles. The study focuses on the fatigue behavior of the composite product and on how it is affected by the branching density of the ethyl groups on the polyolefin backbone and by the winding angle. In two previous papers, the effects of these parameters on the elastic and viscoelastic behavior of the composites was investigated [17], as well as the effects of processing conditions, and gas sterilization on the surface oxidation and cell attachment [18].

2. Experimental

2.1. Materials

Composite materials were produced from Spectra 1000 UHMWPE fibers (Allied Signal) embedded in an ethylene–butene copolymer matrix of the Exact family (ExxonMobil). Three different copolymers were used. Their brochure data are presented in Table 1 [17,19]. The copolymers were supplied in pellets, from which 0.25 mm thick sheets were molded by pressing at 100 °C under a pressure of 6.25 MPa (Carver Laboratory Press), followed by removing them from the press and cooling in an ice-water bath. Filament winding was performed using a bench winder (Burlington Instruments Co., Vermont) as described in Ref. [15]. A flat mandrel (2.5 mm wide, 0.5 mm thick and 135 mm long) was wrapped by a matrix film onto which the fiber was wound at a designated angle, and then wrapped by a second matrix film to produce a preform. The resulting preform was carefully removed from the mandrel and pressed at 100 °C under 22 MPa for 30 min, followed by ice-water cooling. Specimens of three winding angles (the angle between the mandrel axis and the winding direction) of ±28, ±32 and ±42° were produced at a fiber weight fraction of about 0.65. Each specimen is identified by a resin number/winding angle combination, as in Table 2. After pressing, the final strips were 4 mm wide, 0.4 mm thick and 90 mm long.

Table 2
Fatigue test results

Sample	Fatigue limit (MPa)	Fatigue degradation rate (MPa) $(\log N)^{-1}$	Normalized degradation rate $(\log N)^{-1}$
4011/28°	46.4	5.7	0.07
4015/42°	28.3	4.1	0.075
4011/42°	28.4	2.9	0.07
4041/42°	24.6	2.8	0.06

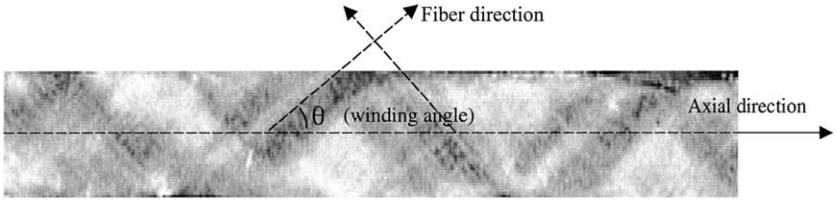

Fig. 1. Schematic presentation of the filament wound strip, indicating the fiber direction and the winding angle.

A scheme of the composite is shown in Fig. 1, while additional experimental details and pictures of the filament wound products can be found in Refs. [17,18].

2.2. Testing

Fatigue tests were performed at room temperature under tension–tension sinusoidal stress control, using a closed loop servohydraulic MTS 858 Mini Bionix testing machine. Load was imposed to ramp to the maximum value at a loading rate of 8 N/s and then to oscillate in a sinusoidal wave form. The minimum to maximum load ratio, *R*, was kept equal to 0.1, and the frequency of the cyclic load was 1 Hz. Samples were gripped by serrated flat face hydraulic grips at a pressure of 10 MPa, in order to avoid slippage during loading. The free length between grips (gauge length) was fixed at 35 mm. In order to compare the results with the previous work [15], the fatigue life was arbitrarily set to an extension limit of 2 mm corresponding to a strain of 5.7%. Tests exceeding 10^6 cycles were stopped even if the 2 mm extension limit was not reached. During the fatigue experiments, signals from the load cell and the LVDT channels were periodically recorded and analyzed in order to determine the load displacement hysteresis loops and maximum specimens elongation. Residual strength measurements were performed on the specimens after fatigue testing, by using the same MTS machine under a constant crosshead speed of 10 mm/min.

3. Results and discussion

The potential biomedical application of the PE fiber reinforced polyolefin composite strip calls for a study of its fatigue resistance and especially of the stress level endured at the conventional fatigue limit of 10^6 cycles. The fatigue behavior summarized by *S–N* curves are presented in Fig. 2 for the three different ethylene–butene copolymer composites. The effect of the winding angle on the fatigue life is demonstrated in Fig. 2 with Exact 4011 for two different winding angles of 42 and 28°. The corresponding data are summarized in Table 2. Because the fatigue life was defined by a demanding criterion for the fatigue strain limit of 5.7% (2 mm extension), corresponding to the static yield strain (see Figs. 9 and 10), most of the specimens did not exhibit any visual damage. Under such conditions, the damage was expected to be fiber independent and to comprise mixed matrix and interfacial shear damage. Hence, a linear regression of the *S–N* curves, incorporating the values of the static yield stress (at 5.7% strain), was appropriate [15], as seen in Fig. 2. The correlation coefficients for the regression lines in Figs. 2 and 3 are high and statistically significant.

The results in Fig. 2 and Table 2 point out the expected similarity of the effects of increasing the branching density of the polymer-matrix and of increasing the fiber angle. Obviously, the strength and stiffness of the composites, which are controlled by these factors, decrease concomitantly [17]. It is noted that the angle effect in the filament wound composites, due to the fiber continuity, is expected to follow the prediction for unidirectional composites loaded at an angle. An important observation is that, selecting a specific combination of copolymer and reinforcement angle can easily regulate the stress level at the conventional fatigue limit of 10^6 cycles, at which the structure is considered fatigue-proof [1]. For the four combinations in Table 2, even the lower values in the range of 25–47 MPa offer a significant safety factor in comparison to the yield point of the ACL (anterior cruciate ligament). For males, mean age of 24.9 years, with unconditioned samples, this has been set at 5 MPa and for quadriceps tendons at 1 MPa. The ultimate strengths of the polyolefin composites are obtained in the range of 400–500 MPa (see Figs. 9 and 10), compared to the ACL of 53.4 MPa and the quadriceps tendons of 33.6 MPa [20].

Because the yield strengths of the four combinations in Table 2 are all sufficiently high, a selection of the best combination should focus on the rate of fatigue degradation.

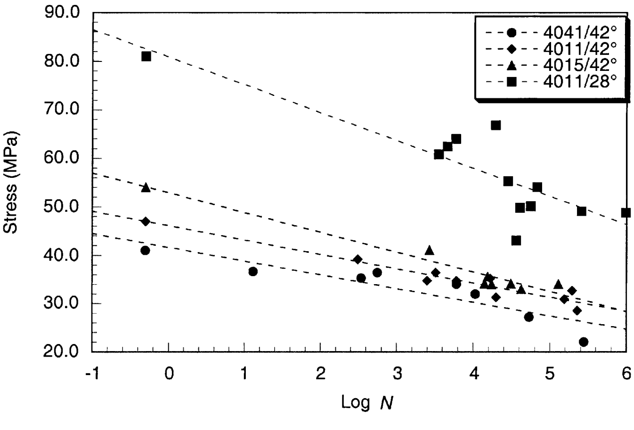

Fig. 2. Fatigue *S–N* curves of three copolymer compositions and two winding angles. The respective correlation coefficients for the regression lines from bottom to top are 0.91, 0.96, 0.98 and 0.86.

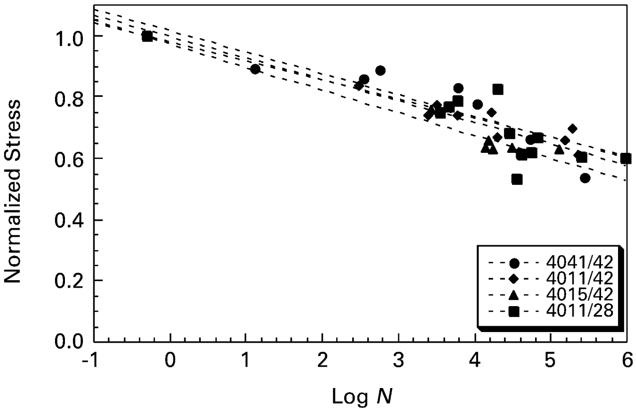

Fig. 3. Normalized fatigue *S–N* curves of three copolymer compositions and two winding angles. The respective correlation coefficients for the regression lines are as shown in Fig. 2.

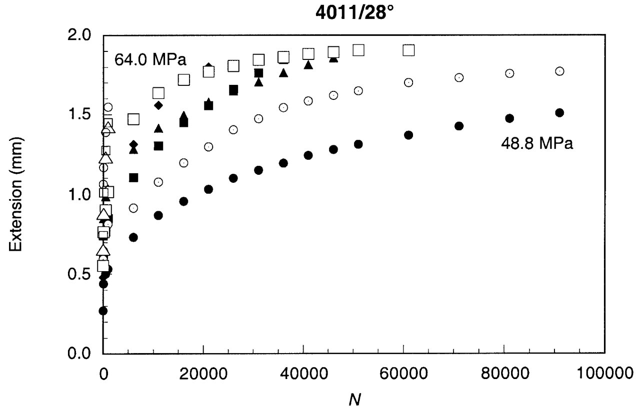

Fig. 5. Fatigue creep extension as a function of the number of cycles up to the predetermined limit of 2 mm extension (5.7% strain) for composites of Exact 4011/28° under stresses in the range of 48.8–64.0 MPa.

The main consideration is to select a combination with the slowest rate of degradation to ensure the highest longevity. Expressing the experimental data in terms of normalized *S–N* plots, in Fig. 3 where the fatigue stress is divided by the static strength of the material can facilitate a comparison on the basis of the degradation rate. It is shown that the rate of degradation decreases with branching and winding angle from 0.075 to 0.060 (log N)$^{-1}$, which is expected to reflect a gradual change in failure mechanism from matrix and interface shear to transverse fracture as the fatigue stress is lowered. Considering the normalized results, it is a significant change hence, for a given stress threshold, higher branching and winding angle are advantageous. This is corroborated by our previous observations that stronger and stiffer composites based on HDPE matrices and higher fiber contents exhibited a significantly lower fatigue limit at 10^6 cycles of 17 MPa [15].

The fatigue experiments can produce additional valuable information pertaining to the designated biomedical applications. This information is about the viscoelastic nature of the composite materials. Because the fatigue experiments were performed under stress control conditions, it was possible to follow the strain increase (fatigue creep) as a function of the number of cycles up to the predetermined limit of 2 mm extension (5.7% strain). The results are demonstrated in Fig. 4 for composites of Exact 4011/42°. The extension is shown for different fatigue stresses in the range of 23.0–36.4 MPa as a function of the number of cycles up to 10^5 cycles, beyond which the change is slow, reaching asymptotically a 2 mm extension. Fig. 5 shows the fatigue creep as a function of the number of cycles for the Exact 4011/28° stiffer composite in the stress range of 48.8–64.0 MPa. The stiffening effect of a smaller angle and lower branching is expected to slow down the creep rate, however, this is not apparent by comparing Fig. 5 with Fig. 4 because of the different fatigue stresses. An overall comparison between the different materials is straightforward by plotting the fatigue creep as a function of the number of cycles for a given fatigue stress. This is shown in Fig. 6 for a fatigue stress of 31 MPa. The branching effect is reflected

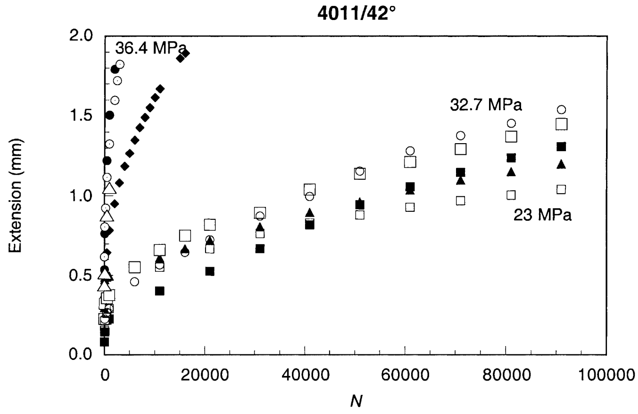

Fig. 4. Fatigue creep extension as a function of the number of cycles up to the predetermined limit of 2 mm extension (5.7% strain) for composites of Exact 4011/42° under stresses in the range of 23.0–36.4 MPa.

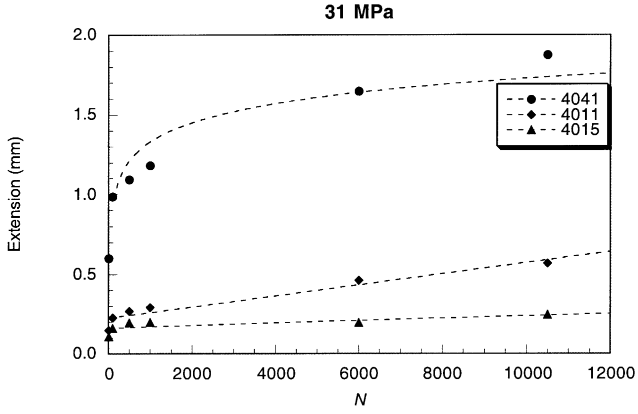

Fig. 6. Fatigue creep extension as a function of the number of cycles, under a fatigue stress of 31 MPa for three copolymer compositions of a winding angle of 42°.

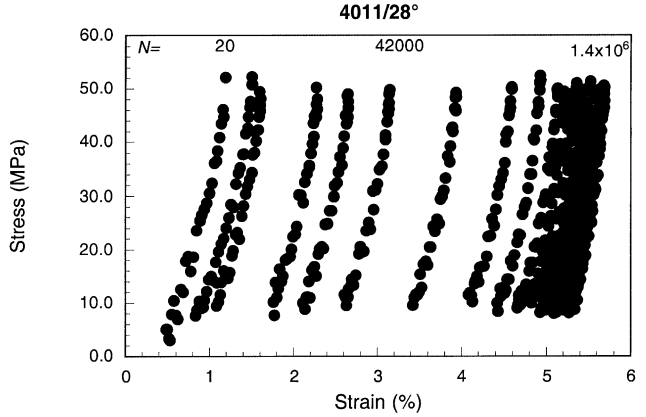

Fig. 7. Hysteresis loops for the Exact 4011/28° composites in the cycle range of 20–1.4 × 10^6.

in the higher extension values obtained already after a small number of cycles.

Another significant method for assessing the viscoelastic nature of the composite materials is based on measuring the viscoelastic energy loss per cycle, termed hysteresis. Figs. 7 and 8 show two sets of hysteresis loops for the Exact 4011/28° and the Exact 4041/42° materials, recorded over a range of fatigue cycles; the serial number of the fatigue cycle, *N*, increases from left to right. Three observations are apparent: The first is that even for a high number of fatigue cycles, the hysteresis loops reflect very low values of energy dissipation per cycle (typical of HDPE [10]). The second observation is that the hysteresis loops are shifted to the right (higher deformation), indicative of the fatigue creep phenomenon discussed earlier, which increases gradually with *N*, approaching the yield strain of the composite material. The third observation is that, unlike the expected fatigue effect [1,14], the modulus increases gradually with *N*, expressing a stiffening effect, which results from fiber realignment in the stress direction.

On the basis of the observations mentioned earlier, it is possible to describe the main features of the fatigue process, being governed, apparently, by the fatigue-generated creep of the matrix. So, the softer composites, those with a higher level of copolymer branching and a higher angle of reinforcement, exhibit slower fatigue degradation rates (the slowest rate is exhibited by the Exact 4041/42° composite). As the deformation approaches gradually the yield strain of the composite, the fibers are forced by the yielding matrix to realign themselves in the stress direction, parallel to the axial direction of the composite strip. This fiber realignment results in a stiffening effect, as the contribution of the fibers to the properties of the composite material becomes more dominant. This two stage behavior, wherein as a result of the fatigue yielding, the fiber's contribution becomes more expressive, can be regarded as a 'second defense line' for the composite strip, being highly desirable in biomedical applications. Moreover, the fiber realignment, which is associated with the fatigue process, ensures sufficiently high residual properties even at the fatigue limit.

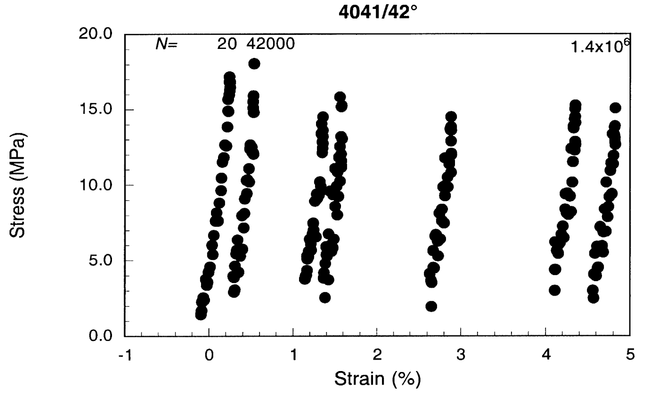

Fig. 8. Hysteresis loops for the Exact 4041/42° composites in the cycle range of 20–1.4 × 10^6.

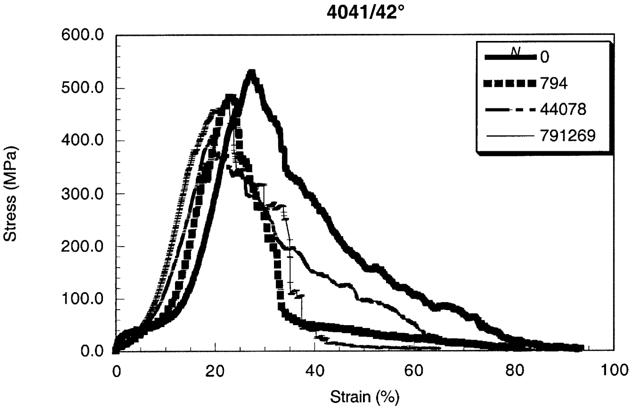

Fig. 9. The residual stress–strain behavior beyond the fatigue limit for the Exact 4041/42° composite, showing the number of cycles endured by each sample at the fatigue limit, and comparing with that of the pristine filament wound composite.

The residual stress–strain behavior beyond the fatigue limit was tested for the Exact 4041/42° and the Exact 4011/42° composites, and a few examples are reported in Figs. 9 and 10, respectively, showing also the number of cycles endured by each sample at the fatigue limit. For each material, the residual stress–strain traces are compared with those of the pristine filament wound composite. In general, the scientific literature [1] predicts the degradation of the mechanical properties of composite materials subjected to fatigue. In our experiments, though, only a minor decrease in the mechanical properties could be detected. The most prominent observation was that the yield point that existed in the pristine material at around 20 MPa disappeared completely, indicating that the fatigue limit of a 2 mm extension correlated with the static yield point of the composite. Therefore, the residual stress–strain behavior reflects the fiber realignment process, which results in retention of the original static properties beyond the yield point. It is seen that the residual stiffness is similar to that of the pristine composite material beyond the yield point, and that the ultimate strength—the so called second defense

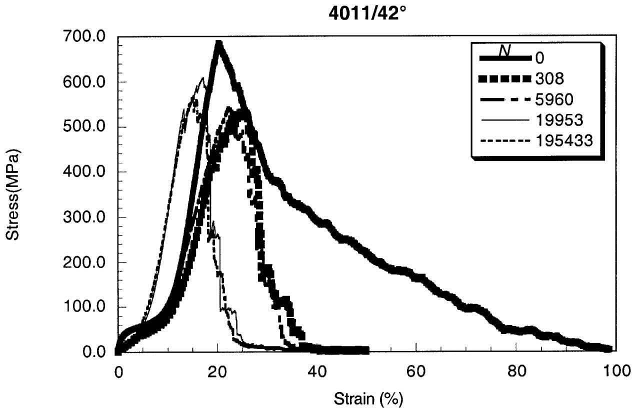

Fig. 10. The residual stress–strain behavior beyond the fatigue limit for the Exact 4011/42° composite, showing the number of cycles endured by each sample at the fatigue limit, and comparing with that of the pristine filament wound composite.

line—which is more than an order of magnitude higher than the yield stress, is reduced only by 10–20%.

4. Conclusions

The fatigue behavior under cyclic loading of filament wound flat strip composites of PE fiber reinforced ethylene–butene copolymer was studied for three different matrix compositions and two winding angles. The matrix composition varied in its ethylene/butene ratio (the level of branching), rendering the composite materials softer and more compliant, as this ratio decreases. A priori, the selection of a material combination for the best fatigue performance is based on meeting a stress level threshold, set by any particular (biomedical) application. Once this prerequisite is satisfied, then, the desirable fatigue life is determined by the fatigue rate of degradation. It was shown that the fatigue rate of degradation decreased as the level of branching and angle of reinforcement increased, so that the more branched copolymers and the higher angles of reinforcement turned out to be more advantageous, with longer fatigue lives. Although the fatigue loading produced yielding and creep, some fiber realignment in the loading direction resulted in significantly high residual properties.

References

[1] Talreja R. Fatigue of composite materials. Tokyo: Techno Publishing Company, 1987.

[2] Strebel JJ, Moet A. Time-dependent fracture toughness measure for polyethylene. J Appl Polym Sci 1994;52(12):1815–21.

[3] Harcup JP, Duckett RA, Ward IM, Capaccio G. Fatigue crack growth in polyethylene: material dependence I: tensile/compressive loading. Polym Engng Sci 2000;40(3):627–34.

[4] Gamstedt EK, Berglund LA, Peijs T. Fatigue mechanisms in unidirectional glass–fiber-reinforced polypropylene. Compos Sci Technol 1999;59:759–68.

[5] Talreja R. Fatigue of composite materials: damage mechanisms and fatigue-life diagrams. Proc R Soc 1981;A378:461–75.

[6] Pegoretti A, Ricco T. Fatigue crack propagation in polypropylene reinforced with short glass fibres. Compos Sci Technol 1999;59:1055–62.

[7] Yeh J, Hong H. Influence of branch length on the dynamic fatigue properties of short-chain branched polyethylenes. Macromol Chem Phys 1995;196:705–14.

[8] Zhou Y, Lu X, Brown N. A fatigue test for controlling the quality of polyethylene copolymers. Polym Engng Sci 1991;31(10):711–6.

[9] Egan BJ, Delatycki O. The morphology, chain structure and fracture behavior of high-density polyethylene. Part I fracture at a constant rate of deflection. J Mater Sci 1995;30:3307–18.

[10] Egan BJ, Delatycki O. The morphology, chain structure and fracture behavior of high-density polyethylene. Part II static fatigue fracture testing. J Mater Sci 1995;30:3351–7.

[11] Kadota K, Chum S, Chudnovsky A. Bridging the PE lifetime under fatigue and creep conditions with its crystallization behavior. J Appl Polym Sci 1993;49(5):863–75.

[12] Martz EO, Goel VK, Pope MH, Park JB. Materials and design of spinal implants—a review. J Biomed Mater Res 1997;38(3):267–88.

[13] Krzypow DJ, Rimnac CM. Cyclic steady state stress–strain behavior of UHMW polyethylene. Biomaterials 2000;21:2081–7.

[14] Kavesh S, Prevorsek DC. Ultra high strength, high modulus polyethylene spectra fibers and composites. Int J Polym Mater 1995;30:15–56.

[15] Shalom S, Harel H, Marom G. Fatigue behavior of flat filament wound polyethylene composites. Compos Sci Technol 1997;57:1423–7.

[16] Pegoretti A, Ashkar M, Migliaresi C, Marom G. Relaxation processes in polyethylene fibre-reinforced polyethylene composites. Compos Sci Technol 2000;60:1181–9.

[17] Kazanci M, Cohn D, Marom G. Elastic and viscoelastic behavior of filament wound polyethylene fiber reinforced polyolefin composites. J Mater Sci 2001;36:2845–50.

[18] Kazanci M, Cohn D, Marom G, Ben-Bassat H. Surface oxidation of polyethylene fiber reinforced polyolefin biomedical composites and its effect on cell attachment. J Mater Sci: Mater Med 2002 in press.

[19] Feigelshtein D, Harel H, Lustiger A, Marom A. Chopped polyethylene fibre reinforced ethylene–butene copolymers. Adv Compos Lett 2000;9:346–53.

[20] Staubli HU, Schatzmann L, Brunner P, Rincon L, Nolte LP. Mechanical tensile properties of the quadriceps tendon and patellar ligament in young adults. Am J Sports Med 1999;1:27–34.