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ELASTIC, VISCOELASTIC AND FATIGUE BEHAVIOR OF POLYETHYLENE FILAMENT WOUND POLYOLEFIN COMPOSITES

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SUMMARY: Filament wound flat strip composites of polyethylene fiber reinforced polyolefins are presented and studied. The versatility of this product is demonstrated by involving different polyolefin compositions based on ethylene-butene copolymers with various winding angles. The dynamic mechanical analysis reflects the viscoelastic nature of the matrix. In particular, the branching density of the polyolefin backbone, expressed by the β-transition peak, dominates the dynamic mechanical behavior. The fatigue results are in agreement with the classical fatigue models, showing that the short-term fatigue behavior, at relatively high stress levels, is 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, is governed by the fatigue rate of degradation, which decreased with the branching density and winding angle.

KEYWORDS: Polyolefin Composites, Filament-Wound Strips, Mechanical Relaxation, Fatigue.

INTRODUCTION

Polyethylene (PE) composites with continuous fiber reinforcement of high volume fraction are known to possess impressive values of specific strength and stiffness. Besides this, different matrix materials and different production techniques, such as filament winding, allow to design composites with unique properties for different applications [1]. It is important to understand the effect of composition and process parameters, such as the matrix type and the angle of reinforcement, on the elastic, viscoelastic and fatigue behavior of these structures.

In filament wound composites, fiber continuity plays a major role in determining the structural performance. Under axial tensile loading, filament wound structures exhibit a yield point followed by a draw region with fiber re-alignment in the loading direction. (In contrast, angle-ply composites usually fail catastrophically by shear [2].) This special case of axial loading of filament wound structures widens their scope of application, offering new engineering and structural properties.

A useful tool for studying the effect of process and product variables on the performance is dynamic mechanical analysis (DMA), being highly sensitive to the inherent properties of the constituents and to their mutual interaction. It is generally accepted that high-density polyethylene (HDPE) exhibits two major transitions while low-density polyethylene (LDPE) exhibits three, including a β transition, attributed to branching. Both HDPE and LDPE exhibit a γ relaxation, occurring between -150°C and -110°C, due to the onset of short-range conformational changes in the amorphous regions and associated with the glass transition. The β relaxation is also thought to relate to the amorphous regions because its magnitude increases with decreasing of the crystalline fraction and the motion of very loose folds and relatively non-extended tie chains [3]. Therefore, it is more marked in LDPE and branched polyethylene below 0°C [4].

Mechanical fatigue is the most common type of failure of composite structures in service [5], where the polymeric matrix develops brittle cracks, which are usually generated by low stresses applied over a long time period [6, 7]. 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 angle ply composites is controlled mostly by the properties of the matrix and of the fiber-matrix interface. The fatigue limit is defined as the stress corresponding to the boundary between the propagating and 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 [8].

The objective of this work is to present and discuss a new type of filament wound, flat strip composites of polyethylene fiber reinforced polyolefins. It follows two preliminary studies by these research groups of the fatigue behavior [2] and relaxation processes [4] 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 elastic, viscoelastic and fatigue behavior of the composite product and on how they are affected by the branching density of the ethyl groups on the polyolefin backbone and by the winding angle.

EXPERIMENTAL

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, whose brochure data are presented in Table 1.

Table 1. Characteristics of the polyethylene-butene copolymer matrices

Matrix	Branching* (per1000 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

*Calculated from the Exact Technical Information

The copolymers were supplied in pellet form, 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 [2]. 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 minutes, followed by ice water cooling. Specimens of three winding angles of 28°, 32° and 42° were produced at a fiber weight fraction of about 0.65. The final strips were 4 mm wide by 0.4 mm thick.

Testing

Axial tensile testing was carried out on an Instron universal tester with a loading gage of 30 mm. The loading rate was 10 mm/min. The modulus and the yield stress in the loading direction x, E_x and σ_x were determined from the initial portion of the stress-strain curve. Tensile dynamic mechanical tests were conducted under liquid nitrogen by a dynamic mechanical analyzer (DMA 983, TA Instruments). All the samples were tested in the temperature range from -150°C to 100°C, at a heating rate of 5°C/min, at a fixed frequency of 1 Hz.

Fatigue tests were performed at room temperature under tension-tension sinusoidal stress control, using a closed loop servohydraulic MTS 858 Mini Bionix testing machine. The minimum to maximum load ratio was kept equal to 0.1, and the frequency of the cyclic load was 1 Hz. The sample gage length was 35 mm. In order to compare the results with the previous work [4], 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.

RESULTS AND DISCUSSION

Axial Tension

Axial tension tests were performed to evaluate the effect of the winding angle on the mechanical properties of the composite. Figure 1 shows the results of the engineering modulus, as a function of the angle θ between the axial and fiber direction.

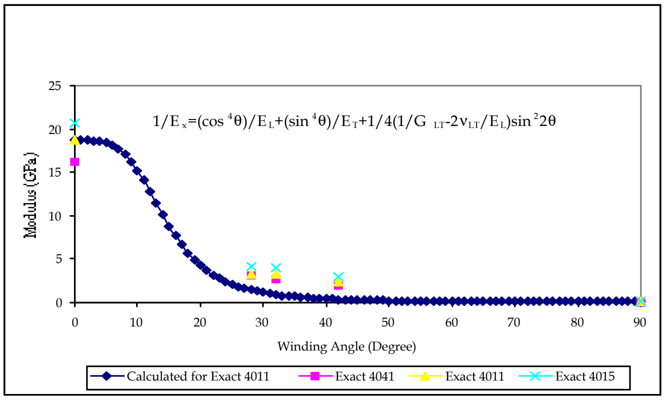


Fig. 1 The engineering modulus as a function of the angle θ between the axial and fiber direction

The experimental results are compared with a theoretical prediction for Exact 4011 composites strips based on a transformation of the principal elastic constants [9]. The compliance along the fiber direction, $1/E_x$, of an orthotropic lamina with its principle axis oriented at an angle θ with respect to the coordinate is expressed as follows:

$$1/E_x = (\cos^4\theta)/E_L + (\sin^4\theta)/E_T + 1/4(1/G_{LT} - 2\nu_{LT}/E_L)\sin^2 2\theta \qquad (1)$$

Where E_x is Young's modulus in the axial direction, E_L, E_T, G_{LT} and ν_{LT} are the Young's moduli, the shear modulus and the Poisson ratio, respectively, of the composite lamina, and L and T donate the principal material axes (longitudinal and transverse).

The strength at an angle θ to the fiber direction is controlled by the mechanical properties of the matrix. Failure occurs by yielding, succeeded by matrix drawing and fiber re-orientation in the loading direction [10]. Figure 2 presents the experimental results of the three systems plotted against θ and compared with the maximum stress criteria for the Exact 4011 composite system.

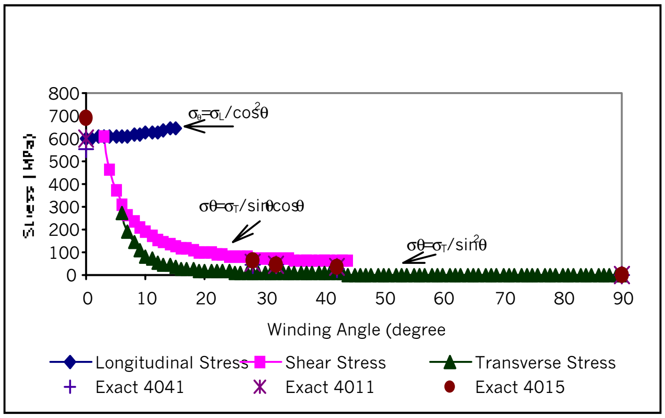


Fig. 2 The ultimate tensile strength as a function of the angle θ between the axial and fiber direction: a comparison with the maximum stress failure criterion.

The values of σ_L, τ_{LT} and σ_T used in the calculations, are the average experimental values obtained for θ = 0, 42 and 90°, respectively.

3.2. Dynamic mechanical experiments

Dynamic tests over a wide temperature range are especially sensitive to the chemical and physical structure of polymers and for studying glass transitions and secondary transitions in amorphous as well as the morphology of crystalline polymers. The real (E') and loss modulus (E'') results display a strong dependence on material parameters such as the branching, winding angle and fiber volume fraction.

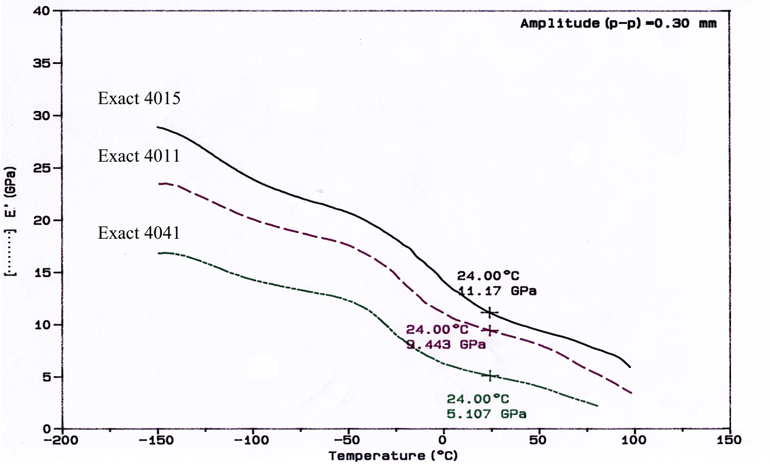


Fig. 3 DMA results of the real modulus for the three copolymers for a constant winding angle and fiber content of 42° and 65%, respectively

As a result, the real modulus at 24°C for a winding angle of 42° increases from 5.1 to 11.2 GPa as the branching of the matrix decreases from 66 to 42 branches per 1000 backbone C atoms (Fig. 3) and to 21.8 GPa as the winding angle is decreased from 42° to 28° (Fig. 4).

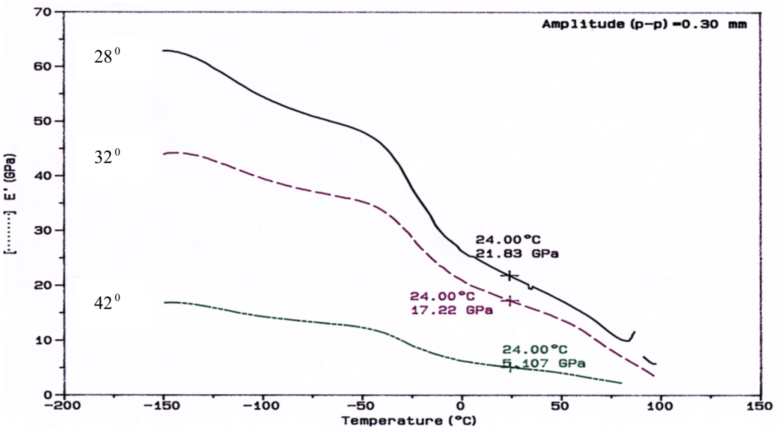


Fig. 4 DMA results of the real modulus for Exact 4041 composites of different winding angles and a constant fiber volume fraction of 65%

These observations are consistent with the theoretical prediction for Young's modulus of composite materials, predicting increased stiffness for higher matrix rigidity and fiber alignment and volume fraction.

The most interesting observation concerns the β relaxation in the temperature range -26°C to -5°C. Although, the origin of this process is still unresolved, it is commonly attributed to motions in the amorphous region. Because in LDPE the β relaxation is a dominant process while in HDPE it is an insignificant one, it has been associated with motion of side chains [11, 12]. This is reflected in the results in Fig. 5 for the copolyolefins, showing a significant β transition in the three copolymers, which is decreasing with the branching density from 1.7 to 1.1 GPa.

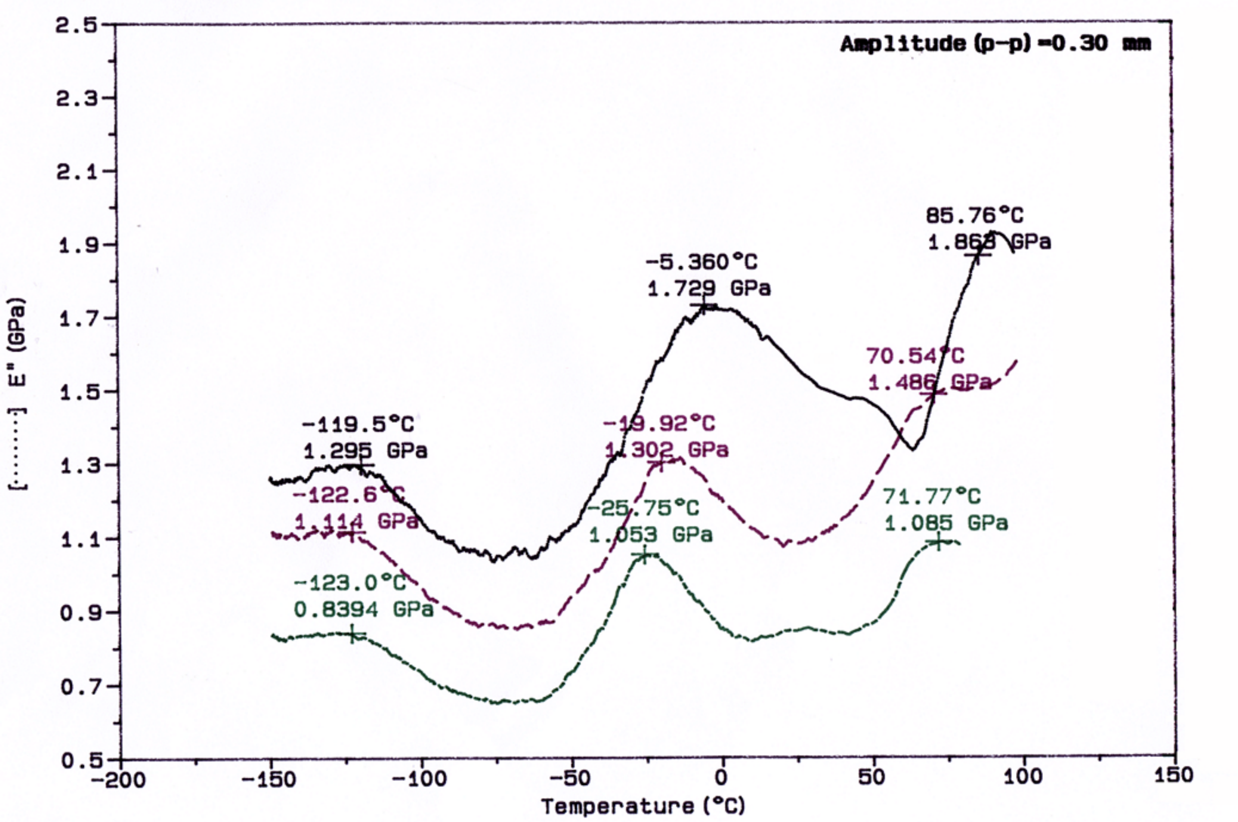


Fig. 5 DMA results of the imaginary modulus for the composites of Fig.3

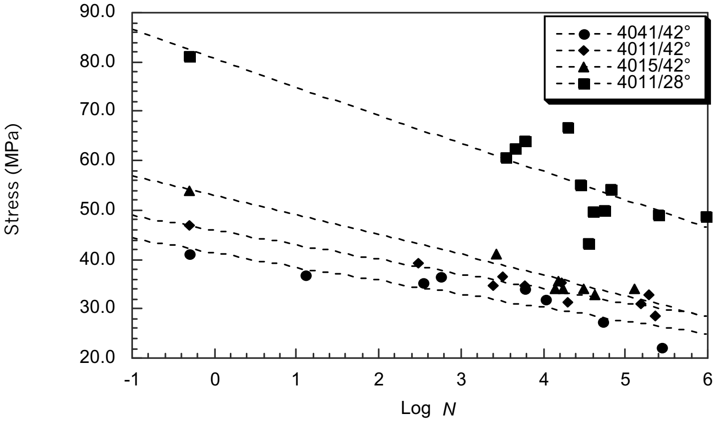
The peak temperature of the β relaxation also reflects the branching density, varying from –5°C to –26°C. In this study, the β peaks shift to higher temperatures with decreasing the ratio of ethylene/butene. It is noted that the relative intensity of the β transition, e.g., the ratio of β/α, does not exhibit a consistent trend with the branching density.

3.3. Fatigue Test Results

The fatigue behavior summarized by *S-N* curves is presented in Fig.6 for the three different ethylene-butene copolymer composites. The effect of the winding angle on the fatigue life is demonstrated with Exact 4011 for two different winding angles of 42^0 and 28^0. The corresponding data are summarized in Table 2.

Table 2. Fatigue test results

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



Normalized degradation rate (log N)$^{-1}$	0.07	0.075	0.07	0.06

Fig. 6 Fatigue *S-N* curves of three copolymer compositions and 2 winding angles

The results in Fig. 6 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. 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 [5]. Because the static 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. The main consideration is to select a combination with the slowest rate of degradation to ensure the highest longevity.

CONCLUSION

This paper presents a new type of filament wound flat strip composite of polyethylene fiber reinforced polyolefins. The versatility of this product is demonstrated by experimenting with different polyolefin compositions based on ethylene-butene copolymers, with a range of fiber volume fractions and various winding angles.

The effects of material parameters, in particular the branching density of the polyolefin backbone and the derived viscoelastic nature of the matrix, on the elastic and viscoelastic behavior of the filament wound composite strip are evident. Dynamic mechanical analysis reveals a significant β transition, which is related to the branching in the copolymer, for the three tested compositions. Increased branching shifts the β transition (and the glass, γ, transition) to lower temperatures and increases the magnitude of the peak. These transitions also reflect the room temperature mechanical properties of the polymer, which are reflected, in turn, in the yield stress and Young's modulus of the composite structure.

It is shown that the fatigue rate of degradation decreases as the level of branching and angle of reinforcement increases, so that the more branched copolymers and the higher angles of reinforcement turns out to be more advantageous, with longer fatigue lives. Although the fatigue loading produces yielding and creep, some fiber realignment in the loading direction results in significantly high residual properties.

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