

Mechanical Properties of PET Short Fiber–Polyester Thermoplastic Elastomer Composites

WUYUN GUO and MICHIO ASHIDA*

The Graduate School of Science and Technology, Kobe University, Nada, Kobe 657, Japan

SYNOPSIS

Presented in this paper is the investigation of the mechanical properties of PET short fiber–polyester thermoplastic elastomer (Hytrel) composites and the discussion of the short fiber reinforcement of the composites. Excellent adhesion of PET fiber to Hytrel elastomer was obtained by treating with isocyanate in toluene solution. The Hytrel composites filled with treated fiber showed a similar tensile behavior, with higher values, to that for the matrix elastomer when fiber loading was less than 5 vol %. The composites loading fibers more than 5 vol % displayed an obvious yield phenomenon, and their yield elongation (between 30–40%) was greater than the fiber's break elongation, which suggested that extensibility of the fiber was quite different from that of the matrix. It is considered that the reinforcement of the short fiber mainly depends on the difference of extensibility between the fiber and the matrix because the difference directly affects the effective transference of the stress from matrix to fiber. The modified parallel model for Young's modulus and yield strength of the composite can be given by the following equations:

$$E_{c0} = \alpha V_f E_{f0} + \beta(1 - V_f) E_{m0},$$

and

$$\sigma_{cy} = V_f \sigma_f(\alpha \varepsilon_y) + (1 - V_f) \sigma_m(\beta \varepsilon_y),$$

respectively, through introducing two effective deformation coefficients, α and β , to represent the extensibility of the fiber and the matrix respectively. The α obtained from the experimental results did not depend on fiber loading but increased with increasing fiber length, and the α for Young's modulus was larger than the one for yield strength, which suggests that α is a function of the strain of the composite and may decrease with increasing the strain, namely, the deformation difference between the fiber and the matrix increases when the strain increases. On the other hand, β is a function of α as:

$$\beta = \frac{1 - \alpha V_f}{1 - V_f}.$$

For the Hytrel elastomer, the maximum of each succeeding stress–strain cycle coincided with the original stress–strain curve for elongations under 600%, but for the Hytrel composites such coincidence was limited to elongations under 30%. This may be caused by the reforming of crystallites in the stress-softened Hytrel elastomer phase at high strain.

© 1993 John Wiley & Sons, Inc.

INTRODUCTION

Elastomer composites reinforced with short fiber combine the elasticity of the matrix with the

strength and stiffness of the loading fiber. The reinforcement of short fiber in the composites is usually manifested as an increase in tensile strength and modulus as well as a decrease in elongation and swelling compared to the matrix elastomer.^{1–9} In a previous paper,¹⁰ we reported on styrenic block copolymers, SIS and SBS, reinforced with PET short fiber, where the composites resulted in an obvious

* To whom correspondence should be addressed.

yield phenomenon and quite different deformation between the fiber and the matrix in extension. The stress of the composites increased by two steps with different modulus under the elongation of the yield point. The tensile properties were influenced by the surface treatment of fiber, fiber length, fiber loading, and matrix type. Hysteresis of the composites was much larger than that of the matrix elastomer and the stress-softening increased with fiber loading and fiber length. These findings suggest that the stress softening of the composites is influenced by the matrix elastomer as well as the loading fiber and that the stress of the composites depends on the hard domain of the matrix at first and then on the soft domain.

Copolyester thermoplastic elastomers (TPE)¹¹ are important engineering thermoplastic elastomers, whose properties are between conventional thermoplastic and conventional thermoset rubber, usually used as replacements for rubber. Different from the styrenic TPE, the copolyester TPE is produced by condensation of dimethyl terephthalate with tetramethylene and polytetramethylene ether glycols; and their hard segments are normally present in the polymer in the form of crystallites distributed throughout a soft, elastic phase.¹² This paper shows the mechanical properties, mainly the tensile properties and hysteresis behavior, for PET short fiber-copolyester TPE composites and elucidates their micromechanics on the basis of short-fiber reinforcement.

EXPERIMENTAL

Materials

The elastomer used was copolyester TPE (DuPont Co., Ltd., trade name Hytrel TR2300, abbreviated as Hytrel); its properties are given in Table I. The short fiber used was polyethylene terephthalate (PET, Teijin Co., Ltd.) with various lengths as given in Table I.

Table I Copolyester Thermoplastic Elastomer, PET Short Fibers, and PET Film Materials

Hytrel (TR2300)	Density (g/cm ³)	1.17
	Hardness (shore A)	32.0
PET fiber	Diameter (mm)	0.028
	Length (mm)	0.5, 2.0, 4.0, 6.0
PET film	Thickness (mm)	0.025

Surface Treatment of PET Fiber

PET fibers were treated by three methods:

1. dipping in a toluene solution containing 5 wt % isocyanate and baking at 115°C for 30 min;
2. dipping in toluene solution containing 5 wt % isocyanate and 4 wt % Hytrel and baking at 115°C for 30 min;
3. treating with O₂ plasma for 5 min, followed by method 1.

Processing

The Hytrel elastomer and short fibers were mixed directly on an open roll with 2-mm clearance. The milling of compounds was always in the same direction in order to orient the fibers along the rolling direction in the sheets. Finally, each stock was passed through the mill and then compressed at 130°C for 15 min to form sheets of 2.0-mm thickness for tensile tests.

Measurement

The PET surfaces after treatment were analyzed with a FTIR 1800 ATR spectrometer (Shimazu Co., Ltd.). The stress-strain curves of the composites were obtained in the direction of the orientation of the fibers at a strain rate of 50 mm/min using an Autograph (Shimazu Co., Ltd.). The section and fracture faces of the composites were coated with sputtered gold and observed by scanning electron microscopy (SEM).

THEORY OF MICROMECHANICS

Mechanical properties of composites in the fiber axis direction can be predicted by a micromechanics approach with the properties of their individual constituents. A good review was made by Abrate¹³ for such an approach. The most important empirical rules for the prediction of modulus and strength of composites were given by Cox,¹⁴ Halpin and Kardos,^{15,16} and Kelly.¹⁷ These empirical rules have been available for the analysis of most short fiber-plastic composites¹⁸ and some short fiber-rubber composites.¹⁹ However, those rules do not explain well the experimental results of most short fiber-elastomer composites in which the matrix has much lower modulus compared with the fiber. This is probably caused by neglecting the characteristics of the high elasticity of elastomer matrix in the composites.

According to the micromechanics analysis for short fiber-reinforced composites,¹³ the applied force transfers from the matrix to the fibers in the zones near the end of the fiber during extension of the composite; the stress is zero at the end of the fiber and increases gradually along the fiber. This results in a low effective stress in the short fiber. On the other hand, the shear modulus of the matrix is critical in developing fiber stress. It is difficult for the short fiber-elastomer composites to develop the fiber stress through the shear stress of the matrix because of the very low shear modulus of the matrix elastomer (about 1/1000 order to one of fiber). Combining elastomer with fiber results in low deformation in the fibers but high in the matrix. Such a conclusion has been proved in the styrenic TPE composites because the matrix always has higher yield elongation than the break elongation of the fiber filled elastomer in extension.¹⁰ Therefore, the reinforcement of short fiber-TPE composites depends not only on the effective stress of the fiber but also, probably to a larger extent, on the transference efficiency of the stress from matrix to fiber, the latter mainly controlled by the different extensibility between fiber and matrix.

The uniaxial tensile stress of the composites at the strain, ε , can be predicted by a parallel model with the stresses of their individual constituents. For short fiber-TPE composites, the deformations of fiber and matrix can be represented by the effective strains, ε_f and ε_m , respectively, which are different from ε . If V_f is the fiber loading, the stress of composite σ_c can be written as:

$$\sigma_c(\varepsilon) = V_f \sigma_f(\varepsilon_f) + (1 - V_f) \sigma_m(\varepsilon_m) \quad (1)$$

where σ_f and σ_m are the stresses at ε_f and ε_m corresponding to fiber and matrix. For convenience, ε_f and ε_m are represented by the ε using two effective deformation coefficients, α and β , that is,

$$\varepsilon_f = \alpha\varepsilon, \quad \varepsilon_m = \beta\varepsilon. \quad (2)$$

α mainly depends on the modulus ratio of fiber to matrix and on fiber length and diameter. Because the modulus ratio is very large, the fiber deformation is smaller than that of the composite and generally $\alpha < 1$. The strain distribution in the matrix phase, as is well known, is uneven for composites and β should be an average deformation coefficient as a macro reflection of the matrix deformation. Thus, we can assume that the deformation of the composite is a sum of both deformations of fiber and matrix described as follows:

$$\begin{aligned} \varepsilon &= V_f \varepsilon_f + (1 - V_f) \varepsilon_m \\ &= V_f \alpha\varepsilon + (1 - V_f) \beta\varepsilon, \end{aligned}$$

then

$$\beta = \frac{1 - \alpha V_f}{1 - V_f}. \quad (3)$$

When ε increases to the yield elongation ε_y , σ_c will reach the yield strength σ_{cy} :

$$\begin{aligned} \sigma_{cy} &= \sigma_c(\varepsilon_y) \\ &= V_f \sigma_f(\alpha\varepsilon_y) + (1 - V_f) \sigma_m(\beta\varepsilon_y) \end{aligned} \quad (4)$$

where $\sigma_f(\alpha\varepsilon_y)$ and $\sigma_m(\beta\varepsilon_y)$ are the stresses of fiber and matrix at each elongation $\alpha\varepsilon_y$ and $\beta\varepsilon_y$, respectively.

The modulus of the composite with respect to the strain of composite is also obtained directly by the differential eq. (1) as follows:

$$\begin{aligned} \frac{d\sigma_c(\varepsilon)}{d\varepsilon} &= V_f \frac{d\sigma_f(\alpha\varepsilon)}{d(\alpha\varepsilon)} \frac{d(\alpha\varepsilon)}{d\varepsilon} \\ &+ (1 - V_f) \frac{d\sigma_m(\beta\varepsilon)}{d(\beta\varepsilon)} \frac{d(\beta\varepsilon)}{d\varepsilon} \\ &= \alpha V_f \frac{d\sigma_f(\alpha\varepsilon)}{d(\alpha\varepsilon)} + \beta(1 - V_f) \frac{d\sigma_m(\beta\varepsilon)}{d(\beta\varepsilon)} \end{aligned}$$

where $d\sigma_c(\varepsilon)/d\varepsilon$, $d\sigma_f(\alpha\varepsilon)/d(\alpha\varepsilon)$, and $d\sigma_m(\beta\varepsilon)/d(\beta\varepsilon)$ are the moduli of the composite $E_c(\varepsilon)$, fiber $E_f(\alpha\varepsilon)$, and matrix $E_m(\beta\varepsilon)$ at the respective deformation. Therefore, the modulus of composite should be equal to:

$$E_c(\varepsilon) = \alpha V_f E_f(\alpha\varepsilon) + \beta(1 - V_f) E_m(\beta\varepsilon). \quad (5)$$

The initial modulus of composite E_{c0} is obtained when $\varepsilon \rightarrow 0$, at the same time because $\alpha\varepsilon \rightarrow 0$ and $\beta\varepsilon \rightarrow 0$, $E_f(\alpha\varepsilon)$ and $E_m(\beta\varepsilon)$ also become the initial modulus E_{f0} and E_{m0} , respectively. Thus E_{c0} is written as:

$$E_{c0} = \alpha V_f E_{f0} + \beta(1 - V_f) E_{m0}. \quad (6)$$

RESULTS AND DISCUSSION

Effect of Bonding

The adhesion between matrix Hytrel and PET fiber was tested with three surface treatments. The tensile strength at the yield point and break elongation are

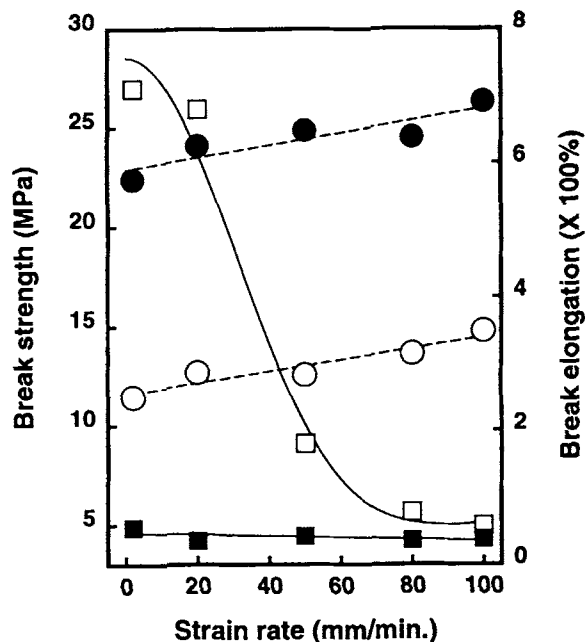
Table II Effect of Fiber Surface Treatment on Break Strength and Elongation for PET Short Fiber-Hytrel Composites

Treatment	Break Strength (MPa)	Break Elongation (%)
1 Without treatment	11.8	185.0
2 Isocyanate in toluene solution	26.7	34.0
3 Isocyanate in Hytrel toluene solution	23.8	39.0
4 O ₂ plasma, then No. 2	23.5	40.0

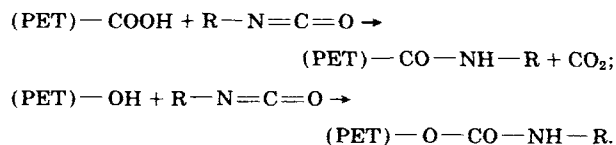
shown in Table II for Hytrel composites loading 10 vol % fiber of 6-mm length. All the composites filled with treated fiber show greater strength and much lower break elongation in comparison with the composites filled with untreated fiber, which indicates that the adhesion between fiber and matrix was improved by the treatments. Table III shows the effect of isocyanate concentration on tensile strength for the composites filled with treated fiber. Although there is no remarkable difference among them, the highest strength was obtained with a concentration of 5 wt %. On the basis of these results, PET fibers were dipped in toluene solution containing 5 wt % isocyanate and baked at 115°C for 30 min.

Figure 1 shows the effect of surface treatment on the break strength and break elongation of the composites at various strain rates. The strength increases linearly with increasing strain rate for both composites filled with treated and untreated fibers, the former having higher values than the latter. On the other hand, the break elongation of the composites with treated fiber decreases only a little with increasing strain rate, but for the composite filled with untreated fiber break elongation falls steeply up to a strain rate of 80 mm/min.

Figure 2 shows FTIR ATR spectra of PET films untreated and treated with 5 wt % isocyanate in toluene solution. The treated film has four absorbance peaks at 3410, 1672, 2900, and 1600 cm⁻¹, considered to be related to —CONH— and —OCONH— groups, respectively. These groups may be produced from the reactions between PET

**Figure 1** Effect of strain rate on break strength (----) and elongation (—) for Hytrel composite. (○, □) untreated and (●, ■) treated by isocyanate bonding agent; 10 vol % fiber of 6 mm length.

molecules end group —COOH or —OH, and isocyanate group —NCO as follows:



The adhesion between the film and the Hytrel matrix is mainly because of the derivative of —CONH— and —OCONH— groups. Strong and medium absorption peaks are found at 2250 cm⁻¹ and 1437 cm⁻¹, respectively, which are ascribed to the —N=C=O— group. This indicates that some isocyanate groups did not react with PET functional groups and are left on the film. However, 5 wt % isocyanate solution seems to be suitable as a bonding agent in the case of the composites because the —COOH or —OH groups exist not only in the molecules of PET fiber but also in the Hytrel matrix.

Table III Effect of Isocyanate Concentration on Break Strength for PET Short Fiber-Hytrel Composites

Concentration (%)	0	1	5	9	17
Strength (MPa)	11.8	23.4	24.6	23.3	21.3

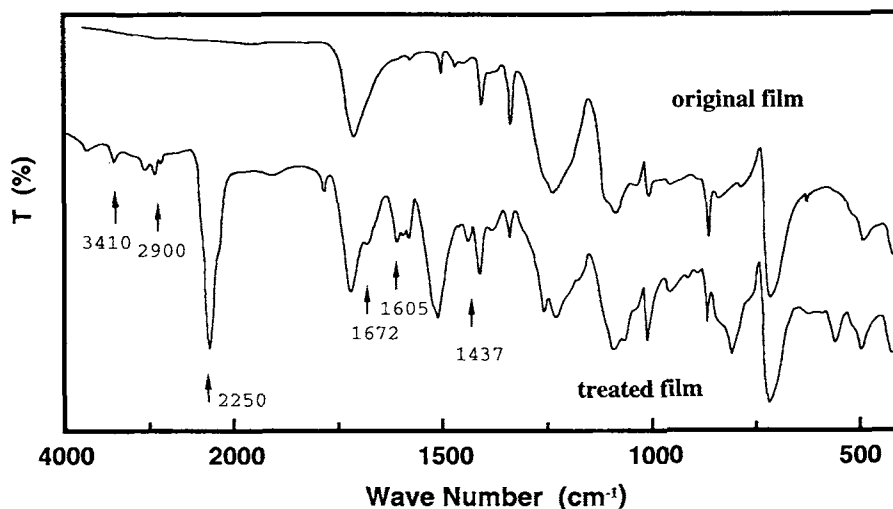


Figure 2 FT-IR ATR spectra of PET films untreated and treated by isocyanate bonding agent.

Figure 3 shows the fracture face for the composite filled with 10 vol % untreated and treated PET fiber 6 mm in length. For the composite with untreated PET fiber in Figure 3(a), some holes where the fibers were pulled out exist on the fracture face, which may be caused by the poor adhesion between fiber and matrix. In the case of the composites with

treated fiber in Figure 3(b), the fracture faces of PET fiber are observed in the photograph and part of the matrix is adhered on the fiber surface. It is therefore evident that the treatment with isocyanate can produce strong adhesion on the interface between TPE matrix and PET fiber. The cross sections of the composites filled with untreated and treated

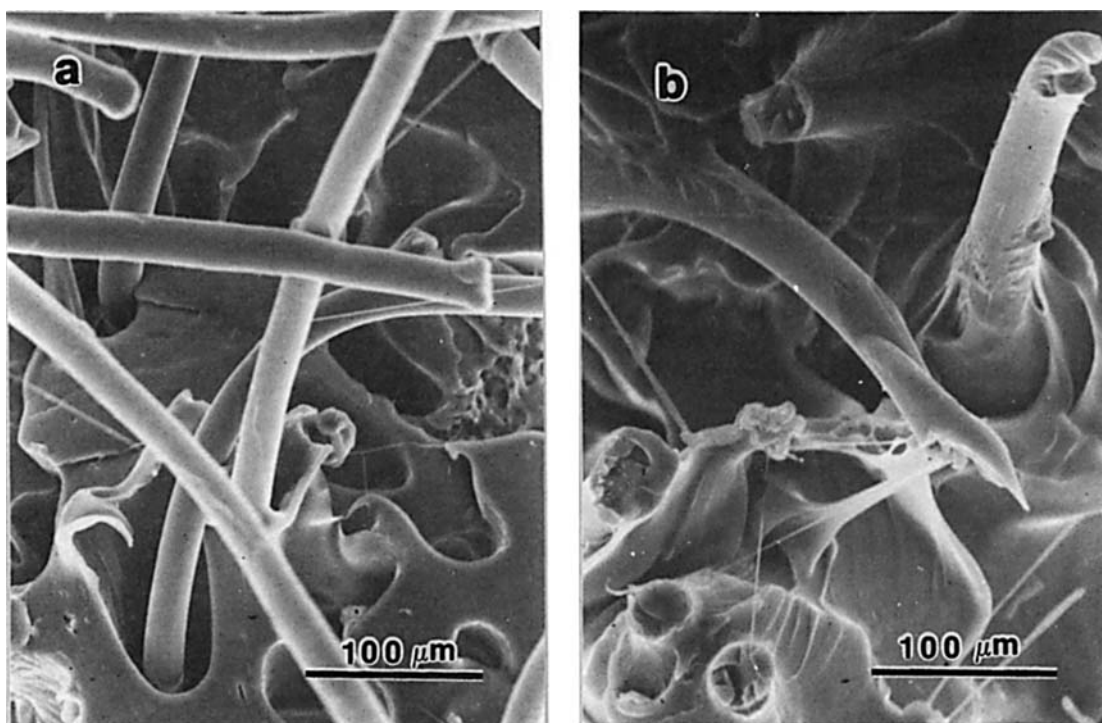


Figure 3 Fracture surfaces of Hytrel composites. (a) Untreated and (b) treated by isocyanate bonding agent; 10 vol.% PET fiber of 6 mm length.

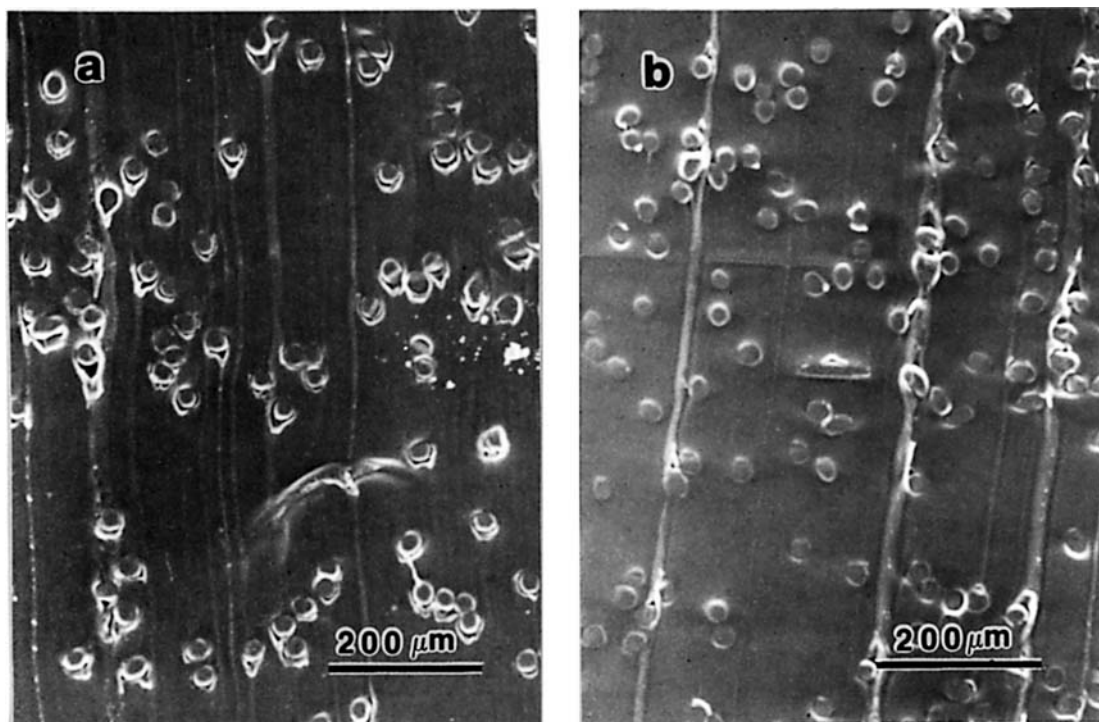


Figure 4 Cross sections of Hytrel composites. (a) Untreated and (b) treated by isocyanate bonding agent; 10 vol.% PET fiber of 6 mm length.

PET fiber are shown in Figures 4(a) and (b), respectively. The figures indicate uniform distribution of PET fibers in the matrix elastomer, but (a) shows gaps between fiber and matrix because of no bonding.

Stress-Strain Behavior

As described for PET fiber-styrenic TPE composites in the previous paper,¹⁰ the stress-strain behavior is controlled not only by the filled fiber but also by the properties of the matrix, and the matrix elastomer undergoes most of the deformation of the composite and the filled fiber absorbs the large internal stress with little deformation during extension.

Figure 5 shows the stress-strain curves for the Hytrel composites containing treated fiber under 10 vol %. The stress increases linearly with increasing strain at small elongation and inflects at about 37% elongation. At strains higher than 40% elongation the curve shows the characteristic sigmoid shape of a rubberlike material with an upward sweep of the curve at strains higher than 600%. When fiber loading is lower than 5 vol %, the stress-strain curves of the composites filled with fiber of 2- and 6-mm lengths show trends closely similar to that of the

matrix elastomer at a little higher stress. The initial linear portion of the composites stress-strain plot becomes steeper with increasing fiber length, but at greater elongation the stress of the composite filled with fiber 6 mm in length has a similar value to that of the composite filled with 2-mm fiber. When fiber loading is 5 vol % and more, the stress-strain curves of the composites show obvious yield phenomena

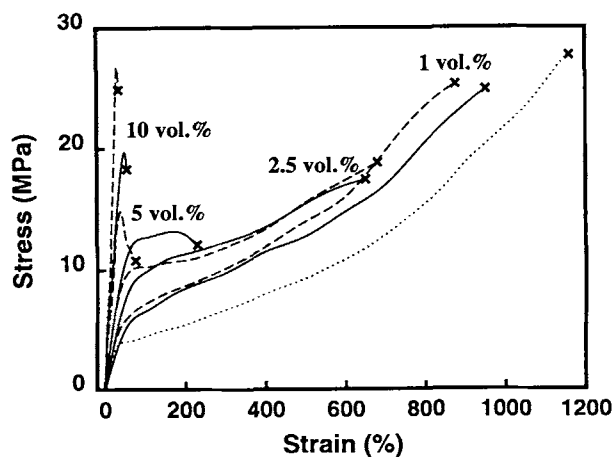


Figure 5 Stress-strain curves for Hytrel (· · · · ·) and its composites containing treated fiber of 2 mm (—) and 6 mm length (---).

and the break elongation becomes shorter with increasing fiber loading. The similarity of the stress-strain curves of the composites with fiber loading under 5 vol % to that of the matrix elastomer suggests that the loading fiber plays a role only as a particulate filler, and that the effect of fiber length on the tensile behavior of the composite may be small if fiber loading is lower than 5 vol %.

Figure 6 shows the effect of fiber loading on the stress-strain curve for the composites filled with treated fiber of 6-mm length. The stress of the composite filled with 5 vol % increases with a little bend with increasing strain, then falls above the yield elongation of about 40%. In the case of the composites filled with fiber of 10 vol % and more, their stresses yield at about 31% elongation near their break points. The modulus and the yield stress of composites increase with increasing fiber loading, but above 10 vol % the increments become smaller. The yield elongation of the composites decreases with increasing fiber loading up to 5 vol %, and then stays almost invariant. Figure 7 shows the effect of fiber length on the stress-strain curve of the composites filled with treated fiber of 10 vol %. The stress-strain curve of the composites loading fiber with length longer than 2 mm shows an obvious yield point and small break elongation different from the composite with fiber of 0.5 mm. The stress of the composite increases and the yield elongation decreases with increasing fiber length. This suggests that the fiber length of 2 mm may be a critical value to achieve effective fiber reinforcement for the Hytrel composites.

The stress-strain behavior for the Hytrel composites also displayed an obvious yield phenomenon

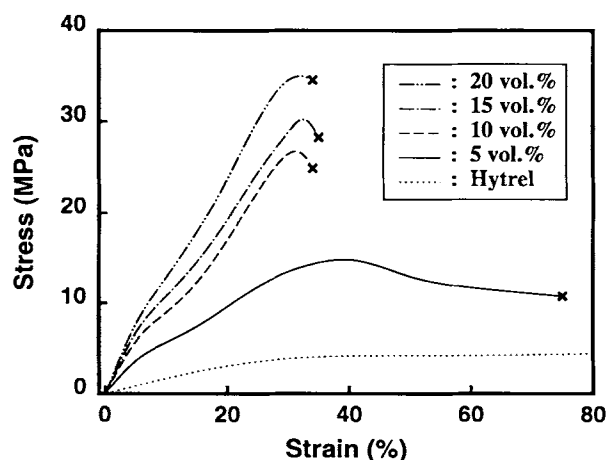


Figure 6 Effect of fiber loading on stress-strain curves for Hytrel composites. Treated fiber of 6 mm length.

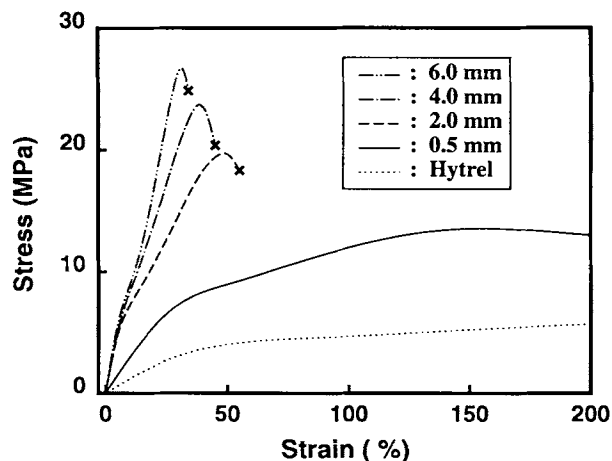


Figure 7 Effect of fiber length on stress-strain curves for the Hytrel composites. 10 vol.% treated fiber.

and longer yield elongation than the break elongation of the fiber, which proved that the fiber deformation was smaller than that of the matrix. In addition, the difference in deformation between fiber and matrix in the Hytrel composites was smaller than that for the styrenic TPE composites because the former TPE has higher modulus than the latter. The yield phenomenon of the Hytrel composites may be caused mainly by separation of fiber from matrix in the zone around the end of the fiber and breaking and restructuring of the matrix at the same zone, just like the styrenic TPE composites.¹⁰

Modulus and Strength

The tensile modulus and strength of the oriented short fiber-elastomer composites varies with fiber loading, aspect ratio, and orientation. The experimental results can be discussed with respect to the modified parallel model derived. The modulus of PET fiber and Hytrel elastomer used in the model were 11.4 GPa and 20.1 MPa, respectively, and the stress-strain data of PET fiber were defined in the previous paper.⁹

All the stress-strain curves for the Hytrel composites had a linear portion from the origin up to strain of about 2%. This portion at low strain was used to determine Young's modulus E_0 , the modulus at zero strain. Figure 8 shows the effect of fiber loading and fiber length on Young's modulus of the Hytrel composites filled with treated fiber. The Young's modulus of the composites filled with the fiber of 4- and 6-mm length increases almost linearly with increasing fiber loading up to 10 or 15 vol %; in the case of the composites filled with fibers shorter than

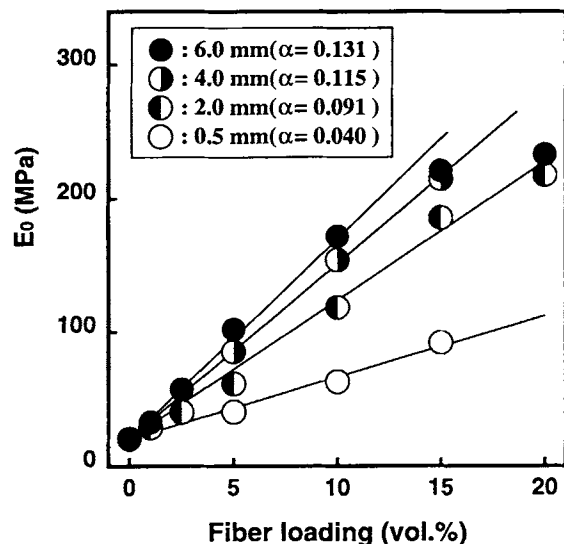


Figure 8 Effect of fiber loading and fiber length on Young's modulus of Hytel composites with treated fiber. The lines were drawn by eq. (6).

2 mm, the Young's moduli increases little with increasing fiber loading, which shows a close resemblance to a particulate composite. As can be seen in Figure 8, the fit of eq. (6) to the experiment is very good for all the fiber lengths. The value of α in the figure is decided by the best fit and obviously, it does not depend on fiber loading but varies monotonously with fiber length. In addition, the Young's moduli are lower than the calculated values when fiber loading is higher than 10 or 15 vol %. The result seems to be attributed to poor fiber orientation in these composites because the mixing and rolling become difficult for those specimens.

Figure 9 shows the effect of fiber loading and fiber length on the yield strength of the Hytel composites containing 10 vol % treated fiber. As well as Young's modulus of the composites, the yield strength increases almost linearly with increasing fiber loading up to 10 or 15 vol %. When fiber loading is more than 10 or 15 vol %, the strength increment becomes smaller. With the various α values corresponding to the fiber length in the figure, the predictions by eq. (4) are good for the yield-strength data [Fig. 9, (—)]. As with Young's modulus, the α of the yield strength does not depend on the fiber loading but increases with increasing fiber length. However, the α for yield strength displays lower values than those for Young's modulus. Therefore, α is a function of the strain of the composite and probably decreases with increasing strain, namely, the difference in deformation between fiber and matrix increases when the strain increases.

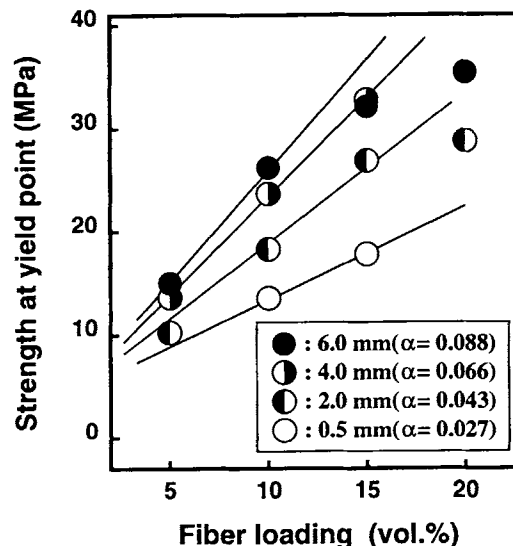


Figure 9 Effect of fiber loading on strength at yield point for Hytel composites with treated fibers. The lines were drawn by eq. (4).

Figure 10 shows the stress at 25% strain, break strength, and elongation as a function of angle θ between the fiber axis and the tensile direction for the Hytel composite containing 10 vol % treated fiber 6 mm in length. The stress at 25% strain and

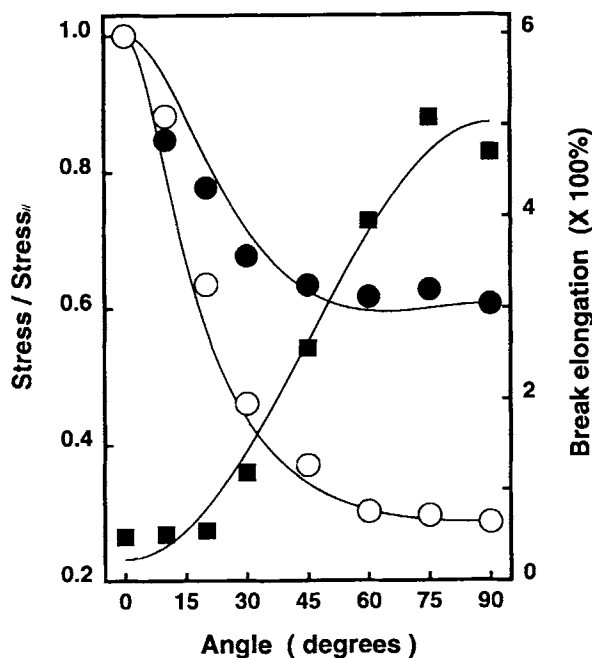


Figure 10 Effect of angle to fiber orientation direction on stress at 25% strain (○), break strength (●) and break elongation (■) for Hytel composite. 10 vol.% treated fiber 6 mm length.

maximum stress falls down rapidly with increasing θ up to 45° , and then the decrease becomes smaller above 45° . In addition, the rate of decrease of the maximum strength is smaller than that of the stresses at 25%. On the other hand, break elongation has little change for small angles of θ , but increases significantly from 20° to 75° .

Hysteresis

When a new sample of the elastomer composite is stretched to a given elongation and then retracted, the subsequent extension to the same strain requires a smaller force. Most of the softening occurs during the first deformation cycle, and after a few stressing cycles a steady state is reached. The stress softening of the Hytrel composites with treated fiber had similar behavior to that of the styrenic TPE composites with treated fiber at the first stress-strain cycle to the given elongation of 35%. The effect of fiber length on the stress softening is shown in Figure 11. The softening increases steeply at fiber length of about 2 mm and increases gradually at lengths above 4 mm. The stress softening of the Hytrel composites during stress-strain cycles is mainly caused by the interface separation around the end of the fiber under large strain and to the breaking and restructuring of hard domain in the matrix as in the styrenic TPE composites.

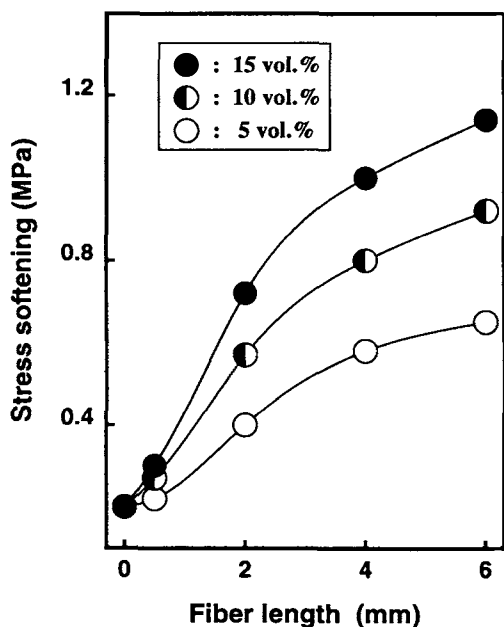


Figure 11 Effect of fiber length and fiber loading on stress softening in the first hysteresis loop for Hytrel composites with treated fiber.

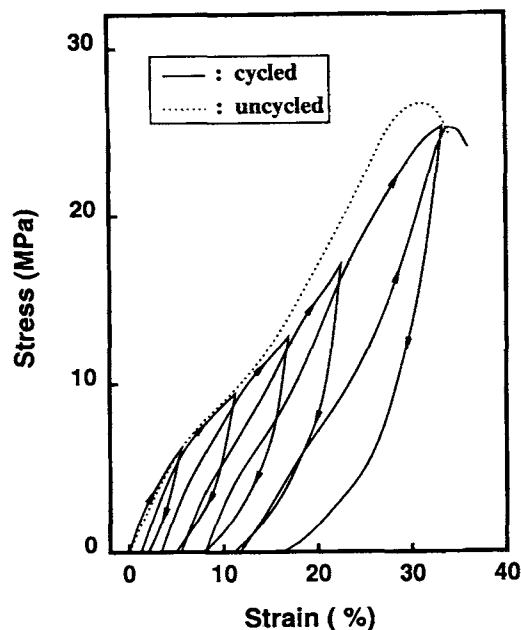


Figure 12 Stress-strain curve and hysteresis loops for Hytrel composites. 10 vol.% treated fiber of 6 mm length.

According to Mullins Effect,^{20,21} when the second and subsequent stressing cycles are examined above the maximum strain reached during the first extension for rubber, the behavior of rubber is similar to that of rubber not previously extended. This effect has been proved in some kinds of filled rubbers.²² Figure 12 shows plots of repeating stress versus strain cycles for the Hytrel composite filled with 10 vol % treated fiber 6 mm in length, for which each succeeding stress-strain cycle is stressed to a higher level in incremental steps. The stress-strain curve of a fresh sample extended to break on the initial stressing (Fig. 12, dashed line). The discrepancy between the maximum stress of the loops and the original stress-strain curve increases with increasing strain. The maximum stresses in the loops for the composites are plotted in Figure 13. The dotted lines are the stress-strain curves for the respective fresh samples. For the Hytrel elastomer, the maximum stress of the loops agrees well with the original stress-strain curve under the extension of about 600%. This may be a phenomenon in which the sample changed from the original translucence to milky white color when the extension was greater than about 600%. Such a change in color of the sample may imply reformation of the crystallites in the sample. Therefore, the disagreement at extensions greater than 600% can be caused by the reformation of the crystallites in the stress-softened Hytrel elastomer. On the other hand, the composite filled with

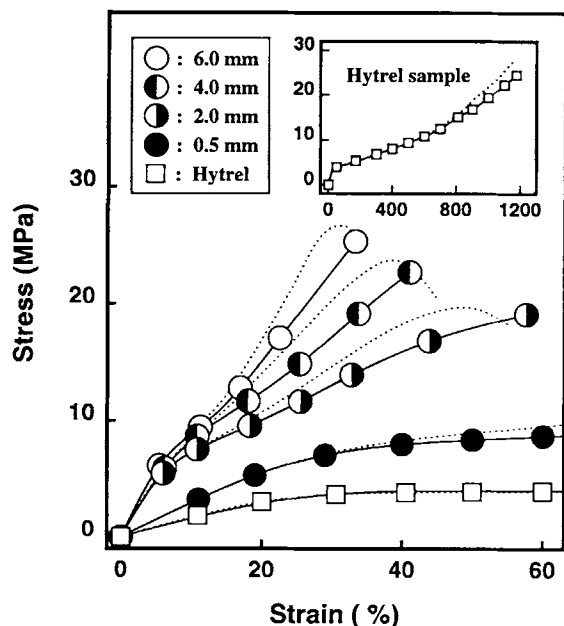


Figure 13 Maximum stress in each hysteresis loop versus strain for Hytrel composites (—) and their original stress-strain curves (· · · · · ·). 10 vol.% treated fiber.

fiber 0.5 mm in length diverges from Hytrel behavior at the lower extension of about 30%. When the fiber length is longer than 2 mm, the discrepancy in stress occurs under about 10% extension. The reformation of the crystallites in the composite occurs first in the high strain area and is reached at a lower extension than that of the elastomer alone because the strain in the matrix surrounding the end of the fiber has a much higher value than the one surrounding the center of the fiber.

CONCLUSION

The PET short fiber-Hytrel TPE composites were prepared and their mechanical properties were discussed for the stress-strain behavior, tensile, and hysteresis properties. The results obtained can be summarized as follows.

1. The PET short fiber-Hytrel TPE composite with high strength can be prepared with the bonding agent of isocyanate in toluene solution that showed excellent interface adhesion for the PET fiber-Hytrel system. The strong adhesion between PET fiber and Hytrel may be attributed to the chemical cross-links, —CONH— and —OCONH—, between PET fiber and Hytrel matrix.

2. When fiber loading was less than 5 vol %, the stress-strain curve of the composites resembled those of the matrix elastomer through a higher value, and the initial increment of the stress was larger with increasing fiber length but the stress at greater elongations did not depend on fiber length. When fiber loading was more than 5 vol %, the composite showed an obvious yield phenomenon, and their yield elongations between 30 and 40% were longer than the fiber's break elongation just like the styrenic TPE composites, which indicated the difference of extensibility between fiber and matrix.
3. By considering the different extensibility between fiber and matrix, the modified parallel model of Young's modulus and yield strength for the composites was modified as follows:

$$E_{c0} = \alpha V_f E_{f0} + \beta(1 - V_f) E_{m0},$$

and

$$\sigma_{cy} = V_f \sigma_f(\alpha \epsilon_y) + (1 - V_f) \sigma_m(\beta \epsilon_y),$$

respectively, through introducing two effective deformation coefficients, α and β . α did not depend on fiber loading but increased with increasing fiber length and strain. β was a function of α as:

$$\beta = \frac{1 - \alpha V_f}{1 - V_f}.$$

4. Discrepancy between the maximum of each succeeding stress-strain cycle and the original stress-strain curve for the composites occurred at lower elongations than that of the Hytrel elastomer, which may be because reformation of crystallites occurs at lower extension because of some high strain matrix area in the composite.

REFERENCES

1. A. Y. Coran, K. Boustany, and P. Hamed, *Rubber Chem. Technol.*, **47**, 396 (1974).
2. A. P. Foldi, *Rubber Chem. Technol.*, **49**, 379 (1976).
3. J. E. O'Connor, *Rubber Chem. Technol.*, **50**, 945 (1977).
4. V. V. Moshev, *Internal. Polymeric Mater.*, **8**, 153 (1980).
5. V. M. Murty and S. K. De, *Rubber Chem. Technol.*, **55**, 287 (1982).

6. L. A. Goettler and S. K. Shen, *Rubber Chem. Technol.*, **56**, 619 (1983).
7. M. Ashida and T. Noguchi, *Composite Polymers*, **2**, 339 (1989).
8. M. Ashida and T. Noguchi, *J. Appl. Polym. Sci.*, **29**, 4107 (1984); **30**, 1011 (1985).
9. M. Ashida, T. Noguchi, and S. Mashimo, *Japan Gomu Kyokaiishi*, **56**, 768 (1983); **60**, 159 (1987).
10. M. Ashida and W. Guo, *J. Appl. Polym. Sci.*, to appear.
11. T. W. Sheridan, in *Handbook of Thermoplastic Elastomers*, 2nd ed., Chap. 6, B. M. Walker and C. P. Rader, Eds., Van Nostrand Reinhold Co., New York, 1988.
12. W. H. Buck, R. J. Cella, Jr., E. K. Gladding, and J. R. Wolfe, Jr., *Morphology and Physical Properties of Polyether-ester Thermoplastic elastomers*, A. S. Dunn, Ed., *J. Polymer Sci.*, Symposium **48**, 47 (1974).
13. S. Abrate, *Rubber Chem. Technol.*, **59**, 384 (1986).
14. H. L. Cox, *Br. J. Appl. Phys.*, **3**, 72 (1952).
15. J. C. Halpin, *J. Elastoplastics*, **3**, 732 (1979).
16. J. C. Halpin and J. L. Kardos, *J. Appl. Phys.*, **43**, 2235 (1972).
17. A. Kelly, and W. R. Tyson, in *High Strength Materials*, V. F. Zackay, Ed., John Wiley & Sons, New York, 1965, p. 578.
18. D. Hull, *An Introduction to Composite Materials*, Chap. 5, and 9, Cambridge Univ. Press, Cambridge, UK, 1982.
19. M. Ashida and T. Noguchi, *J. Appl. Polym. Sci.*, **29**, 661 (1984).
20. L. Mullins, *J. Rubber Res.*, **16**, 275 (1947).
21. L. Mullins, *J. Phys. Colloid Chem.*, **54**, 239 (1950).
22. J. A. C. Harwood and A. R. Payne, *J. Appl. Polym. Sci.*, **11**, 1825 (1967).

Received September 11, 1992

Accepted September 25, 1992