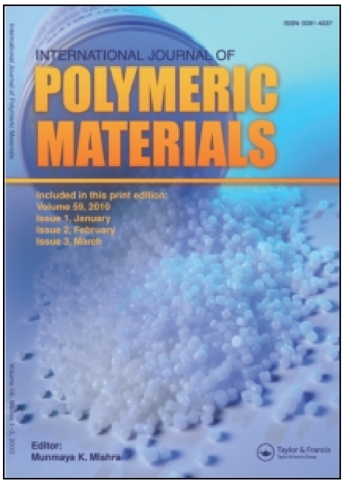

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Mechanical Properties of Short Polyethylene Terephthalate Fiber-Thermoplastic Polyurethane Composite

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The effect of short polyester terephthalate (PET) fiber loading on the mechanical properties of thermoplastic polyurethane has been studied. Tensile strength and tear strength were increased with fiber loading after an initial reduction up to 10–20 phr. Elongation at break was reduced drastically beyond 10 phr fiber loading. Impact strength registered a reduction with increasing fiber content. Anisotropy in mechanical properties was evident beyond 20 phr fiber loading. Tensile strength and tear strength were higher in the longitudinal orientation of fibers than in the transverse orientation of fibers. Scanning electron microscopic studies of the fracture surfaces revealed good correlation with the observed properties.

KEY WORDS Polyethylene terephthalate fibers, thermoplastic polyurethane, composites, mechanical properties.

1. INTRODUCTION

Short fiber-rubber composites combine the strength and stiffness of fiber and the elastic behavior of rubber. Mechanical properties of different short fiber-rubber composites have been reported.[1–5] O'Connor[6] compared the mechanical properties of composites with five kinds of fibers and concluded that the variables like fiber type, fiber content, fiber aspect ratio, fiber orientation, fiber dispersion and fiber-matrix adhesion had a profound influence on the ultimate mechanical properties. The effect of milling parameters on the fiber orientation and hence on the mechanical properties has been reported.[7,8] Early works involved the natural fibers like Jute,[9] Silk[10] and cellulose. Later on synthetic short fibers too found its position

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in rubber composites.[11] The matrices used varied from natural to synthetic and also thermoplastic elastomers.[12] The authors have reported the mechanical properties of short Kevlar fiber-millable polyurethane and -thermoplastic polyurethane composites.[13,14] Rheological and stress relaxation behavior of short Kevlar fiber-TPU have also been reported.[15,16] Dynamic mechanical properties of short fiber-rubber composites have been studied in detail by Ashida and Noguchi.[17-19]

In this paper mechanical properties of short PET fiber-thermoplastic polyurethane (TPU) composite are reported. Emphasis has been given to the effect of fiber loading and fiber orientation on the mechanical properties. Fracture surfaces have been analyzed with scanning electron microscope and an attempt has been made to correlate with the mechanical properties.

2. EXPERIMENTAL

Ether based thermoplastic polyurethane (Estane 58311, molecular weight = 1.5 E5 and $T_g = -21°C$ from DMA) used in this study was obtained from Urethane India and dipped polyethylene terephthalate (PET) cord chopped to 6 mm length (fiber diameter = 21 μ) was procured from Madura Coats, India.

Formulation of the mixes are given in Table I. Kevlar staple fibers and pellets of TPU were dried at 105°C for 2 hours in an air oven. The mixing was carried out in a Brabender plasticorder model PLE 330, fitted with a cam type mixing head, at a temperature of 180°C and at a rotor speed of 60 rpm for six minutes. The mixing sequence is shown in Table II. The molten mass from the plasticorder was sheeted out immediately on an open two roll mixing mill at tight nip. Sheets of 2 mm thickness were molded at 180°C for 3 minutes and quench cooled by immersing the mold in water. Test specimens were punched out along and across the milling direction. Schematic representation of fiber orientation in the test samples is shown in Figure 1. Tensile and tear testing were done as per ASTM D412-

TABLE I

Formulation of the mixes

	Mix No				
Ingredient	TP0	TP10	TP20	TP30	TP40
TPU	100	100	100	100	100
PET	—	10	20	30	40

TABLE II

Mixing sequence

Ingredient	Time, min	RPM	Ram
½ TPU	0	30	up
Fiber	1.5	30	up
½ TPU	3.0	60	down
—	9.0	—	dump

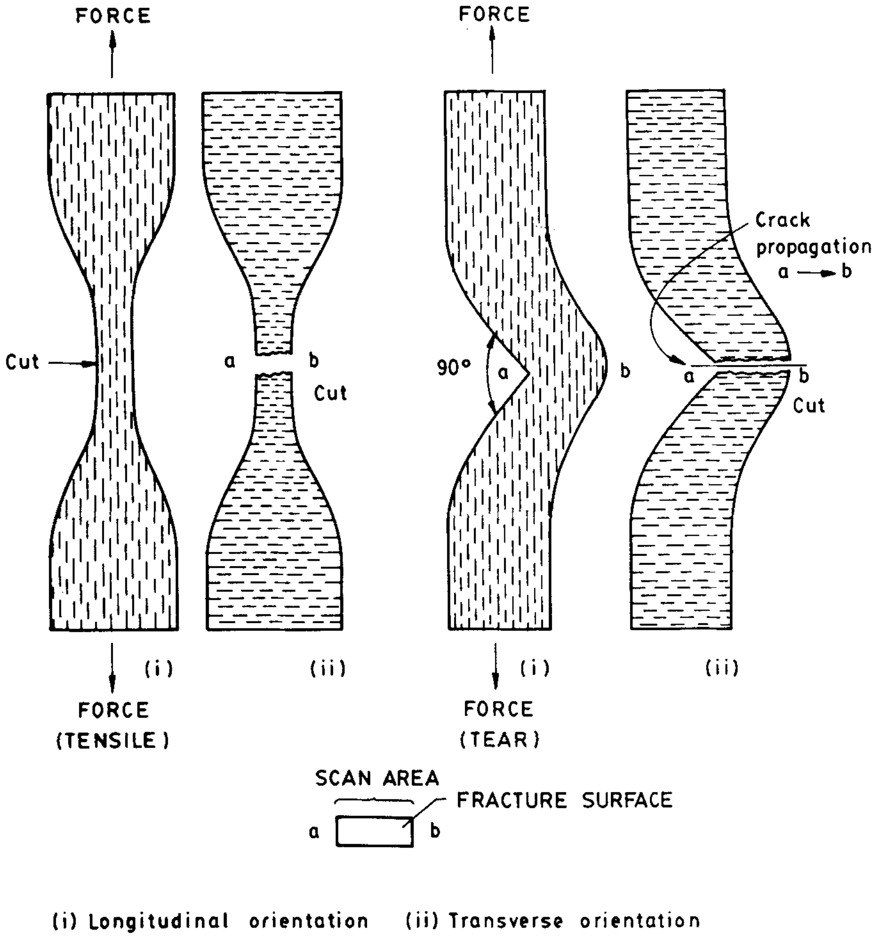


FIGURE 1 Schematic representation of fiber orientation in tensile test sample.

80 and ASTM 624-81 respectively using Instron UTM model 1195. Impact test was carried out on a Ceast Impact tester model 6545/000 as per Din 53448.

3. RESULTS AND DISCUSSION

3.1 Fiber Breakage

Fibers were extracted by dissolving the matrix in tetrahydrofuran. Fiber lengths of a representative sample of about 150 fibers were measured using a traveling microscope and the length distribution was as shown in Figure 2. About 50% of the fibers fall in the region 2–3 mm and about 40% are lower than 2 mm. Fibers longer than 3 mm are very limited. This indicates that the fibers undergo breakage due to shear during mixing in brabender. Czarnecki and White have reported fiber breakage in glass, cellulose and Kevlar fiber-polystyrene system under shear.[20]

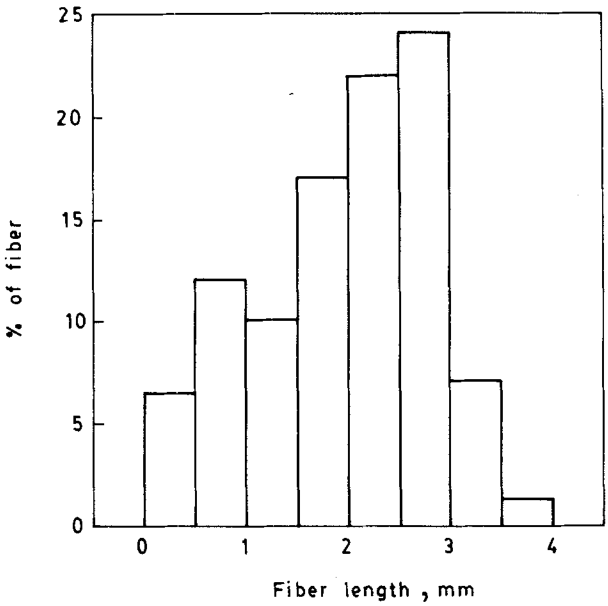


FIGURE 2 Fiber length distribution after mixing.

3.2 Mechanical Properties

Figure 3 shows the stress-strain properties of PET-TPU composite at different fiber loading. Gradual change from predominantly viscoelastic to elastic response is evident from the figure.

It has been reported that mixing in a plasticorder for 4 minutes, sheeting out on a two roll mill and then subjecting the sheet to remixing for 2 more minutes give better fiber dispersion.[12] Table III gives a comparison of the tensile strengths obtained by two modes of mixing. It can be noted that such practice has only a marginal effect on the mechanical properties. So in this study all the samples were mixed continuously for 6 minutes and then sheeted out on a mixing mill.

Table IV gives the mechanical properties of PET-TPU composite at fiber loadings 0–40 phr.

3.2.1. Tensile strength. The variation of tensile strength with respect to fiber loading in both longitudinal (L) and transverse (T) directions is shown in Figure 4. Tensile strength is reduced first in the presence of 10 phr of short PET fibers after which it steeply increases with respect to increasing fiber content in the L direction. At 40 phr of fiber loading the tensile strength becomes equal to that of the unfilled TPU. In the T direction, the tensile strength falls at 10 phr and remains constant with further increase in fiber content. Similar trend in the case of short Kevlar fiber-TPU composite has been reported[14] and also in the case of short cellulose fiber-NR composites.[21] The reduction in tensile strength in the presence of short fibers may be resulting from the following facts. TPU has a segmental multiphase structure consisting of hard and soft segments. The amorphous soft segments are elastomeric in behavior and gives elastic properties to TPU. The hard

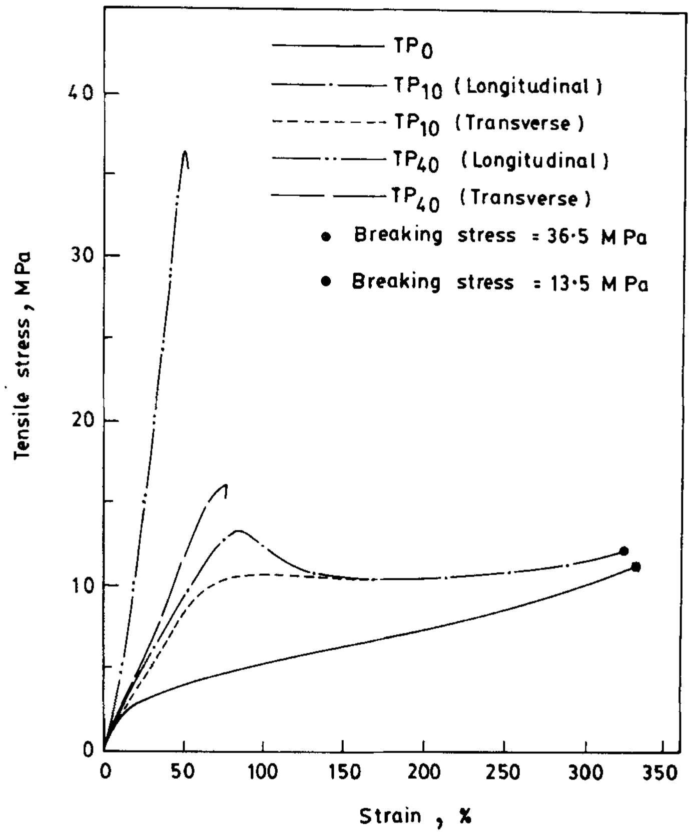


FIGURE 3 Stress-strain behavior at different fiber loading.

TABLE III

Effect of mode of mixing on the tensile strength

Mixing time (minutes)		Tensile strength				
		TP0	TP10	TP20	TP30	TP40
6	L*	36.5	13.5	25.8	29.5	37.1
	T	36.7	13.5	11.5	14.5	15.0
4 + 2	L	36.0	14.2	22.3	31.7	36.6
	T	36.5	15.6	14.4	14.9	14.0

* L = longitudinal and T = transverse orientation of fibers.

segments on the other hand impart the thermoplastic characteristic and act as thermally reversible virtual cross linking and also as reinforcing fillers. In the presence of fibers, there are two effects operating—the dilution of the matrix arising from their physical presence and the reinforcing effect of the fibers. At lower fiber content the number of fibers available for restraining the matrix is lower and hence the dilution effect is predominant, thus lowering the tensile strength. At higher fiber loading, the reinforcing effect of the fiber becomes prominent as

TABLE IV

Mechanical properties of mixes TP0-TP40

Property	Orientation	Mix No TP0	TP10	TP20	TP30	TP40
Tensile strength Mpa	L	36.5	13.5	25.8	29.5	37.1
	T	36.7	13.5	11.5	14.5	15.0
Elongation at break, %	L	590	560	85	45	45
	T	620	480	125	105	75
Modulus at 300% elongation	L	10.0	18.5	—	—	—
	T	10.5	13.5	—	—	—
Tear strength kN/M	L	106	96	97	108	123
	T	105	101	96	90	91
Impact strength $\times 10^{-3}$ J/m	L	>12	1.3	1.7	2.0	2.4
	T	>12	3.4	1.8	1.7	1.8
Hardness, shore A	L	81	89	91	92	92

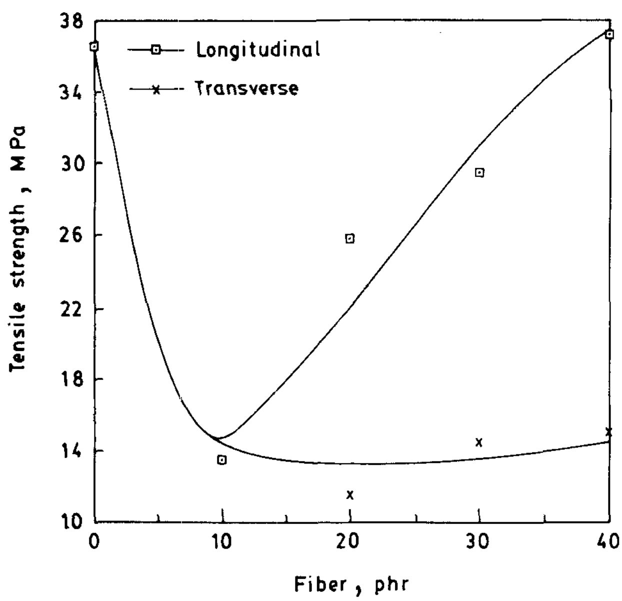
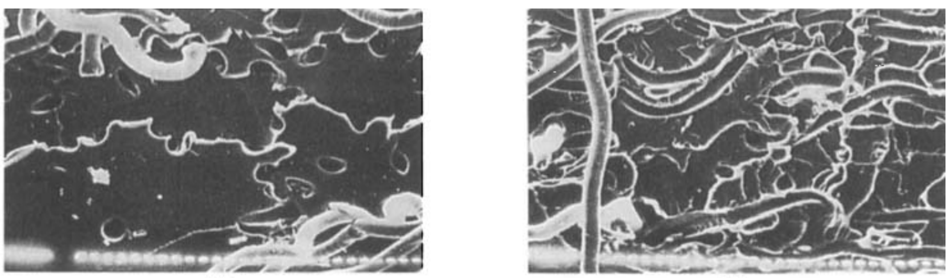


FIGURE 4 Variation of tensile strength with fiber loading.

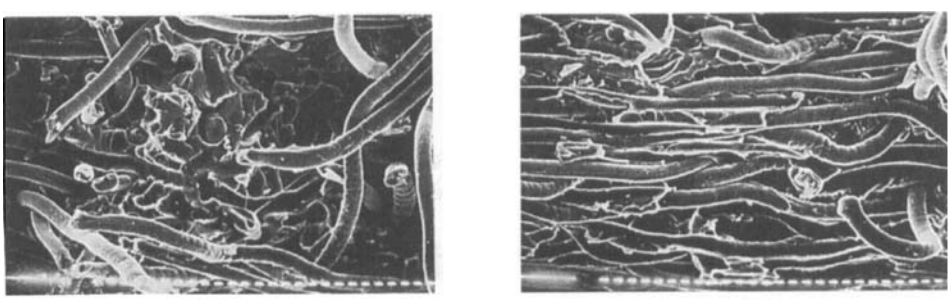
the number of fibers per unit volume is more thus facilitating efficient stress transfer from matrix to fiber. In the L direction, the fibers are able to carry load. Being parallel to the load and at failure, they give effective hindrance to the propagation of the fracture. Whereas in the T direction, being oriented perpendicular to the load, the fibers contribute little to the strength and the fracture propagates easily through fiber-matrix interface, resulting in lower ultimate strength.

SEM analysis of the tensile fracture surface supports this view. Figure 5a shows the SEM photomicrograph of mix TP10 with fiber in the longitudinal direction. High extent of fiber pull out resulting from high interfacial stress is evident from the figure. At low fiber loading, high matrix deformation results in high stress at fiber-matrix interface. The fracture propagation is seen deflected and arrested by the fibers. In this process some of the fibers get pulled out. Preferential orientation of the fiber in the longitudinal direction is evident from the fibers lying normal to the surface. In the transverse orientation, at 10 phr loading, the fibers oriented along the fracture surface are seen (Figure 5b). High extent of interfacial failure is quite clear from the channel left behind by the fibers. Relatively higher matrix deformation compared to longitudinal orientation is also indicated by the figure. At 40 phr, however, the matrix is more restrained by the fibers and hence there is less amount of fiber pull out as is evident from Figure 5c. The layer-like structure seen on the surface indicates a brittle type of failure. The fiber-ends clearly indicates that the fibers have not undergone any microfibrillation as in the case of TPU-Kevlar composite.[14] In the T orientation of fiber at 40 phr, the fracture surface (Figure 5d) shows neatly stacked fibers embedded partly in the matrix. Deformation pattern of the matrix points to a ductile failure which is also supported by comparatively higher elongation at break of the *T* oriented samples. The smooth surface



(a) (b)

FIGURE 5 (a) SEM fractograph of tensile rupture surface of 10 phr fiber filled sample (fibers in the longitudinal direction). (b) SEM fractograph of tensile rupture surface of 10 phr fiber filled sample (fibers in the transverse direction).



(c) (d)

FIGURE 5 (c) SEM fractograph of tensile rupture surface of 40 phr fiber filled sample (fibers in the longitudinal direction). (d) SEM fractograph of tensile rupture surface of 40 phr fiber filled sample (fibers in the transverse direction).

of the fibers indicates absence of kinking during mixing.[20,22] High level of kinking resulting from high shear during mixing in plasticorder has been reported in the case of Kevlar-TPU composite.[14]

3.2.2. Elongation at break. Variation of elongation at break with fiber content is shown in Figure 6. Elongation at break is reduced drastically at 15 phr and beyond 20 phr the effect is only marginal. In the L direction it is lower than in the T direction above a fiber loading of 20 phr as the matrix is more restrained with fibers oriented along the sample length.

3.2.3. Tear strength. Variation of tear strength with increasing fiber content is shown in Figure 7. In the L direction tear strength shows a reduction at 10–20 phr and a linear increase on further increase in fiber loading. With the fibers in the T direction, the tear strength is reduced at all fiber loading studied. Tear strength in the L direction is higher than that in the T direction beyond 20 phr fiber content. In the tear test specimen as the load is applied, the triaxial stress at the 90° angle accumulates which initiates failure at an early stage because of restricted deformation of the matrix in the presence of the fibers. The failure then propagates as strain increases. Any hindrance to the free propagation of fracture will delay tear and hence will improve tear strength. In the L orientation of fibers the propagation of fracture front is effectively hindered by the fibers compared to the transverse orientation of fibers. In the T orientation the fracture easily propagates through fiber-matrix interface as is evident from the SEM studies of the fracture surface.

Figure 8a and 8b show the SEM tear fractograph of mix TP10 in the L and T orientations of fiber respectively. Presence of sinusoidal structure on the fracture is characteristic of high strength matrix and has been reported in the case of other

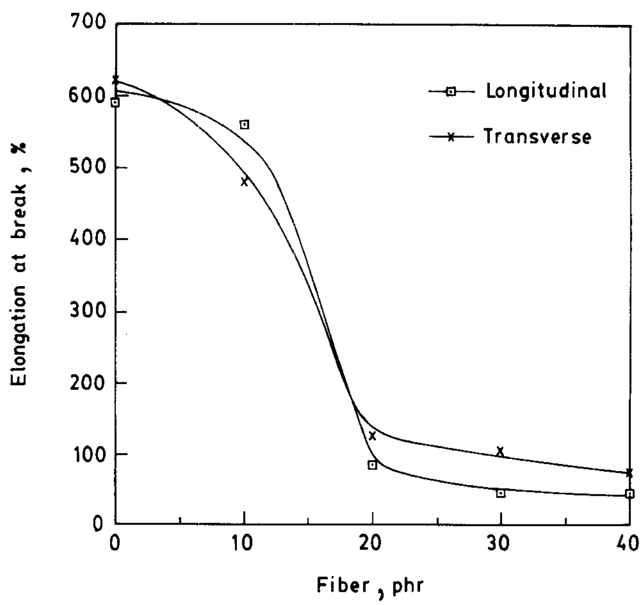


FIGURE 6 Variation of elongation at break with fiber loading.

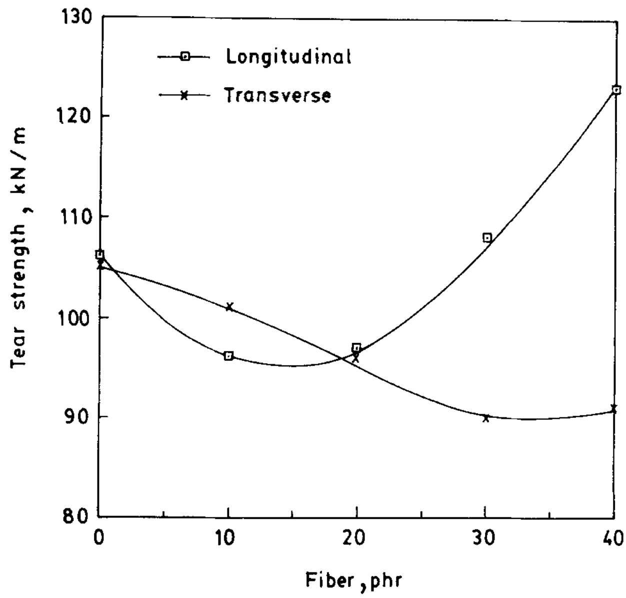
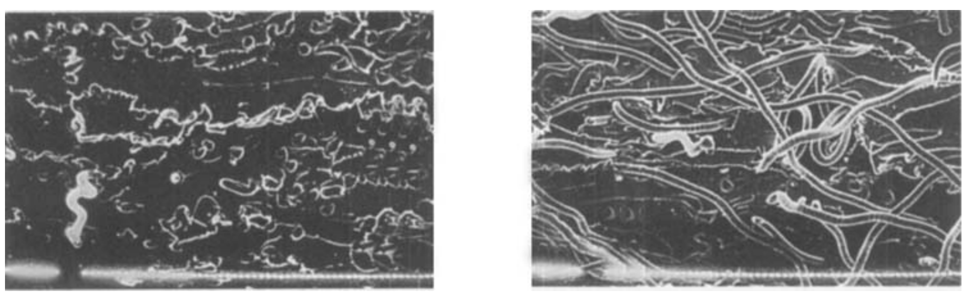


FIGURE 7 Variation of tear strength with fiber loading.

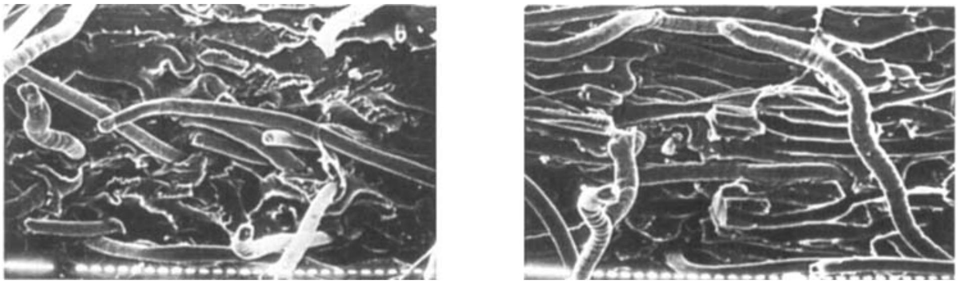


(a) (b)

FIGURE 8 (a) SEM fractograph of tear fracture surface of 10 phr fiber filled sample (fibers in the longitudinal direction). (b) SEM fractograph of tear fracture surface of 10 phr fiber filled sample (fibers in the transverse direction).

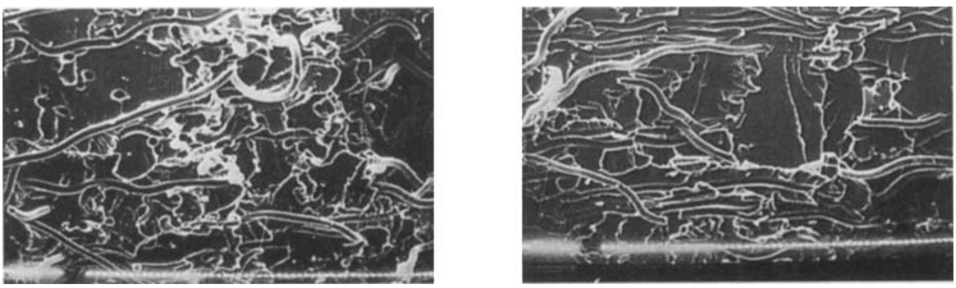
semicrystalline polymers such as 1,2 polybutadiene and thermoplastic copolyester elastomers.[23,24] In the L direction the fracture path is hindered by the fibers and are deflected. High fiber pull out at the initiation region points to high interfacial stress. Similarly in the T direction (Figure 8b) the propagation of tear through the fiber-matrix interface leaves the fiber bare at the surface. Propagation of tear releases the strain energy during which some of the fibers are hooped, as seen in the figure. Tear fracture surfaces of 40 phr fiber loaded samples are shown in Figure 8c and 8d in the L and T directions respectively. Fiber pull out is limited in the Figure 8c than in the case of low fiber loaded sample (Figure 8a). Sinusoidal patterns are seen here also though to a very limited extent. In the T direction (Figure 8d) fiber-matrix interface failure is prominent.

3.2.4. Impact strength. Impact strength is reduced drastically in the presence



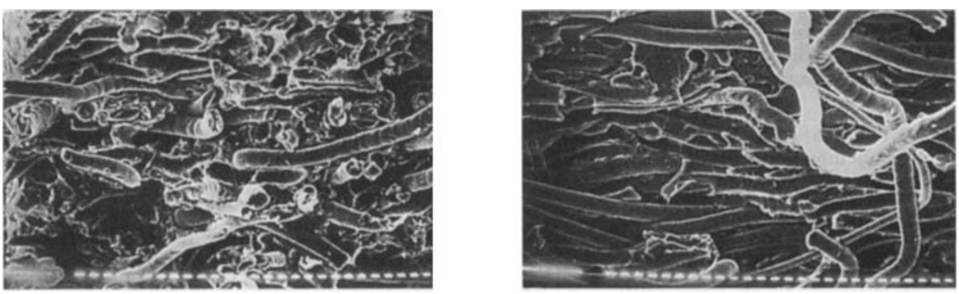
(c) (d)

FIGURE 8 (c) SEM fractograph of tear fracture surface of 40 phr fiber filled sample (fibers in the longitudinal direction). (d) SEM fractograph of tear fracture surface of 40 phr fiber filled sample (fibers in the transverse direction).



(a) (b)

FIGURE 9 (a) SEM fractograph of impact fracture surface of 10 phr fiber filled sample (fibers in the longitudinal direction). (b) SEM fractograph of impact fracture surface of 10 phr fiber filled sample (fibers in the transverse direction).



(c) (d)

FIGURE 9 (c) SEM fractograph of impact fracture surface of 40 phr fiber filled sample (fibers in the longitudinal direction). (d) SEM fractograph of impact fracture surface of 40 phr fiber filled sample (fibers in the transverse direction).

of short PET fibers in both L and T orientations. Impact strength of the unfilled stock is very high owing to its self reinforcing nature. The matrix yields under stress absorbing the energy of impact. But in the presence of fiber, the elongation is restricted and during testing the specimen is subjected to deformation far exceeding the ultimate elongation. Under the impact load the failure is initiated at the fiber-matrix interface which then leads to catastrophic failure. Impact strength in the L direction is higher than in the T direction.

SEM fractograph of the impact failed sample surface of mix TP10 are shown in Figure 9a and 9b with fibers in L and T direction respectively. Smooth holes and looped fibers are seen in the L direction. In the T direction fibers are seen ejected out at one end and with the other embedded in the matrix indicating a sudden impact force trying to pull the fiber off. Fracture is found to propagate in all direction. At 40 phr fiber loading the fracture surface indicates a more restrained matrix and highly disrupted fibers spread on the surface in both L and T directions (Figure 9c and 9d). Matrix seen in Figure 9d indicates that the failure is more of ductile in nature compared to Figure 9c.

Hardness is increased from 81 to 91 at 10 phr and then for further increase in fiber loading up to 40 phr the hardness values remain more or less constant.

4. CONCLUSIONS

The effect of incorporation of short PET fiber in TPU on the mechanical behavior can be summarized as:

Tensile strength shows a minimum at 10 phr beyond which it increases linearly. Tear strength shows a minimum in the range of 10–20 phr of fiber content. Impact strength is reduced drastically with fiber content. Anisotropy in properties is evident above 20 phr with fiber oriented in the longitudinal direction showing better properties. Scanning electron microscopy studies of the fracture surface shows good correlation with the observed properties.

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