Plastics, Rubber and Composites Processing and Applications 19 (1993) 105-110

Effect of processing parameters on the mechanical properties of short Kevlar aramid fibre-thermoplastic polyurethane composite

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(Received 28 August 1991; revised version received 25 February 1992; accepted 27 February 1992)

Abstract: The effect of various processing parameters, such as nip gap, friction ratio and roll temperature, on the tensile properties of short Kevlar aramid fibre-thermoplastic polyurethane composite has been investigated and the tensile and tear fracture surfaces have been characterised using a scanning electron microscope. A nip gap of 0.45 mm, a friction ratio of 1.15 and a roll temperature of 62°C was found to give optimum mechanical properties. Scanning electron microscopy study revealed a higher extent of fibre orientation in the milling direction in the above condition.

1 Introduction

Short fibre-rubber composites combine the strength and stiffness of the fibres and the elastic behaviour of rubber. The mechanical properties of these composites were the focus of earlier studies.¹⁻⁵ Early works involved the natural fibres like jute,⁶ silk⁷ and cellulose. Later on, synthetic short fibres also found their position in rubber composites.8 The matrices used varied from natural through synthetic to thermoplastic elastomers.9 O'Connor¹⁰ compared the mechanical properties of composites with five kinds of fibres and concluded that the variables such as fibre type, fibre content, fibre aspect ratio, fibre orientation, fibre dispersion and fibre-matrix adhesion had a profound influence on the ultimate mechanical properties. The authors have reported the mechanical properties of short Kevlar aramid fibre filled millable polyurethane and thermoplastic polyurethane.^{11, 12} (Kevlar is a Du Pont registered trademark.) Rheological and stress relaxation behaviour of short Kevlar aramid fibre-thermoplastic polyurethane (TPU) composite have also been reported.13, 14

In the case of short fibre-elastomer composites, the fibres are oriented preferentially in one direction by sheeting out on a two-roll mixing mill. The effect of various milling parameters on the fibre orientation in the case of short cellulose fibre-natural rubber composite has been dealt with by Moghe.¹⁵ He pointed out that the maximum extent of fibre orientation in the milling direction was about 60-70%. Kutty and Nando have reported that passing twice through tight nip gave the optimum property to short polyethylene terephthalate fibre-natural rubber composite.¹⁶ In this paper the effect of various milling parameters like nip gap, friction ratio and roll temperature on the mechanical properties of short Kevlar aramid-TPU composite is reported. Tensile and tear fracture surfaces have been characterised by using scanning electron microscopy (SEM).

2 Experimental

Ether based thermoplastic polyurethane (Estane 58311) used in this study was obtained from Urethane India (Madras, India) and Kevlar aramid short fibres (T-970) of length approximately 6 mm and length to diameter ratio 500 were procured from Du Pont De Nemours and Co. (Wilmington, DE, USA).

TPU with 20 phr of short Kevlar aramid fibres were used in this investigation. Kevlar aramid staple fibres and pellets of TPU were dried at 105°C for 2 h 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 6 min. The mixing sequence is shown in Table 1. The mixed stock from the plasticorder was transferred immediately to a two-roll mixing mill and sheeted out at friction ratios ranging from 1.05 to 1.75, at nip gaps ranging from 0.45 to 1.48 mm, and at roll temperatures ranging from room temperature to 100°C. Sheets (2 mm) were compression moulded taking care to avoid disruption of fibre orientation due to flow inside the mould. Tensile and tear specimens were punched out and tested on an Instron universal testing machine as per ASTM D 412-80 and D 624-73 (Die C). Tensile and tear fracture surfaces were sputter coated with gold and examined under SEM. Fibres were extracted by dissolving out the matrix in a solvent and lengths were measured using a travelling microscope. The length distribution was calculated from a representative sample of 80-100 fibres. A schematic representation of the fibre orientation in different test samples is given in Fig. 1.

Table 1. Mixing	1 sequence
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Table 1. Mixing sequence									
Ingredient	Time (min)	Rotor speed (rpm)	Ram						
1/2 TPU 0 Fibre 1⋅5 1/2 TPU 3⋅0 — 9⋅0	30 30 60 —	Up Up Down Dump							
1	naximum e) gʻdirection ido have t u nip ga <u>ve</u>	d out that the <i>x</i> i in the millio Kutty and Nan rice through tigh	te pointe mentation 50-70%. oasstrg tv						
	Crack propagatic	n viti 1							
	a b Cut	$\mathbf{a}_{1}^{[1]} \mathbf{b}_{1}^{[1]} \mathbf{b}$	a						
L温记旧 三三 (i) Force (tear)	EI (ii)	(i)							
Scan area	acture surface	Longitudinal orientation	Transverse orientation						

Fig. 1 Schematic representation of fibre orientation in tensile and tear test specimens.

3 **Results and discussion**

Figure 2 gives the length distribution of fibres after mixing. The average initial length of the fibres was approximately 6 mm. During mixing the fibres break down to smaller lengths. Nearly 50-60% of the fibres are in the 0.5-1.5 mm range after mixing. The high reduction of fibre length can be attributed to the high shearing force they were subjected to during mixing in the Brabender plasticorder.

3.1 Effect of nip gap

Table 2 shows the effect of different nip gap settings on the mechanical properties of short Kevlar aramid-TPU composite. The tensile strength in the longitudinal orientation of fibres shows a gradual reduction from 32 to 27 MPa as the nip gap is increased from 0.45 to 1.48 mm. For transverse



Table 2.	Effect	of	nip	gap o	on the	tensile	properties
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Property	Fibre orien-	Nip opening (mm)						
	tation	0.45	0.63	0.91	1.21	1.48		
Tensile strength (MPa)	Lª T	32 13 (2·5) ^b	31 14 (2·4)	28 15 (2·0)	28 15 (1·9)	27 16 (1·7)		
Elongation at break (%)	L T	12 105	19 72	16 71	19 80	18 79		
Tear strength (kN/m)	L T	77 65	75 66	73 63	74 51	75 54		

 ${}^{a}L =$ longitudinal and T = transverse orientation of fibres. ^bValues in parentheses are anisotropy index values; defined as a ratio of the tensile strength in the longitudinal direction to that in the transverse direction.

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orientation of fibres, however, the tensile strength is increased marginally with increasing nip gap. Tensile strength in the transverse direction is lower than in the longitudinal direction at all nip gaps. The anisotropy index, defined as the ratio of a property in the longitudinal direction to that in the transverse direction, is also given in parentheses in Table 2. The anisotropy index of tensile strength shows a reduction with increasing nip gap. This may be due to the lower extent of fibre orientation at higher nip gaps. Fibres become oriented preferentially in the direction of milling because of the high shear rate at the nip. The shear rate at the nip is defined by the differential roll speed and nip gap. As the nip gap increases at a constant roll speed, the shear rate and hence the extent of fibre orientation decreases, leading to a lower anisotropy index. This view is supported by fracture surface analysis. Figures 3(a)-(c) show the SEM photomicrographs of tensile fracture surfaces at different nip set-ups. Figure 3(a) shows the SEM tensile fractograph at a nip gap of 0.45 mm with the fibres oriented in the longitudinal direction. The fibres projecting out of the matrix and the fibre pull-out holes seen on the fracture surface testify the preferential orientation of fibres in a direction perpendicular to the fracture surface. At the same nip gap, the tensile fractograph of the sample with transverse orientation of fibres is shown in Fig. 3(b). The high extent of orientation of fibres in one direction is clearly evident from the figure. This also explains the observed lower tensile strength in the transverse direction. Unlike the fibres in the longitudinal orientation, in the transverse direction, the fibres are oriented parallel to the direction of the propagating tear. In the longitudinal direction, the majority of fibres are in a direction perpendicular to the direction of propagation of fracture, and they can arrest or deflect the advancing tear, thereby delaying the failure. In this process the stress concentration at the fibre-matrix interface becomes so high that the fibres are pulled out of the matrix on failure. The holes left behind by the pulled-out fibres are seen in Fig. 3(a). It has been reported that the Kevlar aramid fibres undergo kinking during mixing because of high shear in the plasticorder. These fibres may then break at the kinked portions during tensile or tear failure. A detailed study of this has been reported elsewhere.¹² In the transverse direction, the fibres, being oriented parallel to the direction of the advancing tear, are not effective in giving hindrance to or arresting the propagation of fracture. The tear propagates through the fibre-matrix interface, exposing the fibres, as seen in Fig. 3(b). This results in a lower strength in the transverse orientation of fibres.

Fibre orientation is affected by the nip setting, as is evident from Fig. 3(c). The SEM fractograph of a sample with fibres in the transverse direction at a nip



Fig. 3 SEM photomicrographs of tensile fracture surface at: (a) nip gap 0.45 mm, fibres in longitudinal direction; (b) nip gap 0.45 mm, fibres in transverse direction; (c) nip gap 1.48 mm, fibres in transverse direction.

gap of 1.48 mm is shown in Fig. 3(c). Comparison with the fibre orientation at 0.45 mm nip setting (Fig. 3(b)) clearly establishes that a greater number of fibres become oriented in the milling direction at a lower nip gap.

Elongation at break shows a slight increase at higher nip gap in the longitudinal orientation of fibres, whereas in the transverse direction the trend is the reverse. This is because at lower nip gap more fibres are oriented in a direction parallel to the direction of application of force in the longitudinal

samples. Since the elongation of these short fibres is very small compared to the matrix material, the failure is initiated at much lower elongations; but as the nip increases there are a smaller number of fibres to restrict matrix deformation, resulting in higher elongation. In the transverse direction, however, the reverse is true, i.e. when more and more fibres are oriented in a direction perpendicular to the direction of deformation the restriction due to the fibres is less, leading to higher elongation before the failure is initiated. As the fibre orientation in the transverse direction decreases at higher nip, random orientation puts some of these fibres in the longitudinal direction which restricts the matrix deformation in that direction. This will reduce the elongation at break at high nip in the transverse orientation of fibres.

Tear strength in the longitudinal direction remains more or less constant and decreases in the transverse direction with increasing nip gap. This again points to a lower level of fibre orientation at higher nip spacing. The tear strength in the transverse direction is lower than in the longitudinal direction at all nip settings. SEM photomicrographs of the tear fracture surface at two different nip gaps (0.45 and 1.48 mm) in the transverse orientation of fibres are shown in Figs 4(a) and (b). A higher level of fibre orientation at lower nip spacing is evident from the figures.

3.2 Friction ratio

Table 3 gives the variation of tensile properties with increasing friction ratio at a constant nip of 0.45 mm. Tensile strength shows only a marginal increase from 30 to 32 MPa when the friction ratio is increased from 1.05 to 1.15, and it gradually declines on further increasing the friction ratio. A friction ratio of 1.05 is insufficient to orient the fibres to optimum level because of the lower shear rate. At very high shear rates, because of the viscoelastic nature of the matrix, the actual shear to which the bulk is subjected is lower because of probable slip on the roll surface. This again leads to insufficient fibre orientation, resulting in lower tensile strength at very high shear rate. SEM study of the fracture surface is in agreement with this observation. Figures 5(a) and (b) show the fracture surfaces at friction ratios 1.15 and 1.6 with fibres in the longitudinal direction. Protruding fibre ends in Fig. 5(a)clearly indicate higher fibre orientation at a friction of 1.15 compared to 1.6 (Fig. 5(b)). Figure 5(b) indicates a random orientation of fibres. In the transverse direction the tear strength variation with respect to increasing friction ratio is negligible.

Elongation at break remains more or less independent of the friction ratio in the longitudinal direc-





Fig. 4 SEM photomicrographs of tear fracture surface at: (a) nip gap 0.45 mm, fibres in transverse direction; (b) nip gap 1.48 mm, fibres in transverse direction.

(b)

Table 3. Effect of friction ratio on the to	ensile properties
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Property	Fibre	Friction ratio							
	orien- tation	1.05	1.15	1.30	1.45	1.60	1.75		
Tensile strength (MPa)	ect Lhe The	30 15	32 14	30 15	29 15	29 15	26 15		
Elongation at break (%)	Do L og T	29 70	13 105	26 102	26 112	28 94	26 100		
Tear strength (kN/m)		75 80	78 70	76 78	64 96	69 64	61 59		

L = longitudinal and T = transverse orientation of fibres.

tion, and in the transverse direction it is increased at higher friction ratios.

Tear strength shows a maximum of 78 kN/m at a friction ratio of 1.15 below and above which the tear strength is lower. This indicates that the friction ratio for the optimum tensile and tear strength is about 1.15 in the short Kevlar aramid fibre-TPU composite. The SEM fracture surface of a longitudinally fibre oriented tear sample at a friction ratio of 1.15 is









Fig. 5 SEM photomicrographs of tensile fracture surface at: (a) friction ratio of 1.15, fibres in longitudinal direction; (b) friction ratio of 1.60, fibres in longitudinal direction; (c) friction ratio 1.15, fibres in longitudinal direction.

shown in Fig. 5(c). Highly mutilated matrix and projected fibres are seen along with fibre pull-out holes. The advancing fracture front may be disturbing the fibre alignment at the time of tear propagation.

3.3 Roll temperature

Table 4 gives the tensile properties at roll temperatures ranging from 30 to 100°C. Here again, the

Table 4. Ef	Effect	of	roll	temperature	on	the	tensile
				roperties			

Property	Fibre	R	Roll temperature (°C)					
	orien- tation	30	62	86	100			
Tensile	L	32	32	34	35			
strength (MPa)	Ŧ	13	14	15	15			
Elongation	L	12	20	19	19			
at break (%)	Ŧ	105	100	90	93			
Tear	L	64	68	67	68			
strength (kN/m)	Ŧ	63	65	65	67			

L = longitudinal and T = transverse orientation of fibres.

changes are only marginal. The tensile strength increases to 35 MPa from 32 MPa in the longitudinal direction. The higher roll temperature during sheeting of the mixed stock from the plasticorder improves the chances of fibre orientation, thereby increasing the tensile strength. With cold rolls there is the possibility of the stock becoming cooled on rolls on contact with the roll, leading to improper fibre orientation. At higher roll temperatures, the mix remains soft and hence the fibres become oriented at the shear rate in the nip. Elongation at break increases above 30°C in the longitudinal direction and decreases marginally in the transverse direction. SEM study of the fracture surface supports this view. Figure 6(a) shows the tensile fractograph of samples sheeted at 100°C in the longitudinal and transverse directions. A comparison with the corresponding fracture surface at 30°C (Fig. 3(a) confirms the view that a higher roll temperature during sheeting increases the fibre orientation. The tear strength in the longitudinal direction is improved from 64 to 68 kN/m at 62°C roll temperature and remains constant at higher temperatures. In the transverse direction the tear strength is lower than that in the longitudinal direction and increases marginally above 30°C roll temperature. A SEM photomicrograph of the tear fracture surface with fibre in the longitudinal direction at 100°C is shown in Fig. 6(b). It shows a better orientation compared to that at $30^{\circ}C$ (Fig. 6(c)).

4 Conclusions

From the foregoing study the following conclusions may be drawn:

 Processing parameters like nip gap, friction ratio and mill roll temperature have a profound influence on the fibre orientation and hence on the mechanical properties of short Kevlar









Fig. 6 SEM photomicrographs of tensile fracture surface at:
(a) roll temperature 100°C, fibres in longitudinal direction; (b) roll temperature 100°C, fibres in longitudinal direction; (c) roll temperature 30°C, fibres in longitudinal direction.

aramid fibre-thermoplastic polyurethane composite.

- A nip opening of 0.45 mm, a friction ratio of 1.15 and a roll temperature of 62°C give optimum mechanical properties.
- SEM analysis of fracture surfaces supports a high level of fibre orientation at high shear at the nip.

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