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Sunhemp fibre-reinforced polyester

Part 1 *Analysis of tensile and impact properties*

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This paper describes the tensile and impact behaviour of polyester composites reinforced with continuous unidirectional sunhemp fibres of plant origin. The tensile strength and Young's modulus of sunhemp fibre were found to be 389 MPa and 35.4 GPa, respectively. Tensile strength of composites containing up to 0.4 fibre volume fraction (V_f) were found to increase linearly with V_f and the results showed good agreement with the rule of mixtures. The work of fracture, as determined by Izod impact test, was also found to increase linearly with V_f and the work of fracture for 0.24 V_f composite was found to be approximately 21 kJ m^{-2}. The analysis of various energy absorbing mechanisms during impact fracture showed that fibre pull out and interface fracture were the major contributions towards the high toughness of these composites. The results of this study indicate that sunhemp fibres have potential as reinforcing fillers in plastics in order to produce inexpensive materials with a high toughness.

1. Introduction

Ligno-cellulosic based natural fibres are relatively inexpensive and these renewable resources, which are abundantly available, have the potential to be fillers and reinforcements in polymers [1]. The economics of using natural fibre-reinforced polymer composites in developing countries have been highlighted in a previous study [2].

All ligno-cellulosic based natural fibres consist of cellulose microfibrils in an amorphous matrix of lignin and hemicellulose. These fibres consist of several fibrils which run all along the length of the fibre (Fig. 1a): each fibril exhibits a complex layered structure made up of a thin primary wall encircling a thicker secondary layer [3] as shown schematically in Fig. 1b, and is similar to the structure of a single wood fibre [4]. The secondary layer is made up of three distinct layers: the middle layer is by far the thickest and the most important one in determining the mechanical properties of the fibre. In this layer parallel cellulose microfibrils are wound helically around the fibrils and the angle between the fibre axis and the microfibrils is termed the microfibril angle. Natural fibres are themselves cellulose fibre-reinforced materials and the microfibril angle and cellulose content determine the mechanical behaviour of the fibre.

Sunhemp or sunn-hemp (*Crotalarea juncea*) is widely cultivated in India and several other countries [5]. The fibre is extracted from the bast of the plant by a process known as retting and the source and maturity of the plant and the method of retting govern the properties of the fibre [5]. The high cellulose content (70 to 88%) [6] and low microfibril angle (9.8°) of sunhemp fibres as compared to other common natural fibres [7] indicate that sunhemp has potential as a reinforcing filler in polymers.

The flexural behaviour of natural fibre polymer composites such as coir, straw and jute fibres in polyester have been studied and analysed by various workers [1, 8, 9]; however, there has been little work on the work of fracture and the analysis of the various energy absorbing mechanism during impact of these natural fibre composites. In this investigation, the potential of sunhemp–polyester composites in terms of their tensile and impact properties has been evaluated. In this study, the toughness of sunhemp–polyester composites has been analysed in detail, and an attempt is made to understand the complex nature of composite fracture.

2. Experimental procedure

Sunhemp fibres, 1 to 2 m long and 40 to 100 μm diameter, were obtained from local sources. These fibres were rinsed in detergent to remove dirt sticking to the fibres and dried at room temperature for a week.

Tensile testing of sunhemp fibres was carried out on an Instron testing machin at a cross-head speed of 0.5 mm min^{-1}. Specimens were prepared by mounting single fibres on a stiff cardboard piece with a 50 mm window. The ends of the fibres were glued on to the cardboard with epoxy resin. The diameter of the fibre specimens was measured, using an optical stereomicroscope with a graduated eye piece, at five different places along the gauge length and the average cross-sectional area was calculated by assuming that the fibres are cylindrical in shape. Some error is likely to result since these fibres are highly irregular in shape. A total of 25 fibres were tested in tension.

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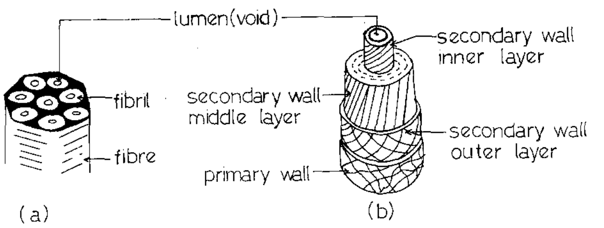

Figure 1 Schematic diagram of (a) a sunhemp fibre and (b) an individual fibril.

Unidirectionally aligned "continuous" sunhemp–polyester composites with varying fibre volume fractions (V_f) were prepared using two slightly different methods. A modified lay-up method was used for low V_f composites and a pultrusion technique for higher V_f composites. In the former technique, sunhemp fibres were cut to fit the length of the mould and the desired amount of fibres corresponding to the desired V_f were weighed and kept aside. General purpose polyester resin was then thoroughly mixed with 1 wt % cobalt naphthonate (accelerator) and 1 wt % methylethylketone peroxide (catalyst). A thin layer of resin mixture was poured into the bottom of the mould and sunhemp fibres were placed over it. This procedure was repeated until all the resin and fibres were utilized. A cover plate was placed over the specimen and a slight pressure was applied. The volume fractions were calculated from the weight fractions and the densities of the fibre and cured polyester resin. The apparent density of sunhemp fibre varied from 1020 to 1149 $kg\,m^{-3}$ and an average of 1075 $kg\,m^{-3}$ was used for calculating the V_f. The apparent density of fibres was measured according to the procedure described by White and Ansell [1].

Specimens containing up to 0.24 V_f were prepared according to the technique described above. However, beyond 0.24 V_f it was not possible to make samples with the modified lay-up technique without application of high pressure. Specimens containing more than 0.24 V_f were, therefore, prepared by the pultrusion technique. This involved soaking the fibres in the resin bath containing 1 wt % accelerator and 1 wt % initiator. The fibres were then pulled through an opening into a mould. In both cases the samples were cured at room temperature for 24 h and then post-cured for 6 h at 80° C.

Tensile test samples prepared by the modified lay-up technique (low V_f) were of dumb-bell shape and dimensions were as recommended by ASTM D638 Type I. Samples prepared by the pultrusion technique were 220 mm long, 12.7 ± 0.3 mm wide and 3.5 ± 0.25 mm thick. End tabs (45 mm long) were glued by epoxy on either side of the pultruded samples. Tensile testing of composites was carried out on an Instron testing machine at a cross-head speed of 0.5 $mm\,min^{-1}$ and the extensometer was used to record the strain. Five samples were tested at each V_f.

Impact test specimens were 65 mm long, 12.7 mm wide and 6 ± 1 mm thick. A notch was cut at the centre of the edge face. The notch size and testing procedure were as given in ASTM D256 Type A. Five samples were tested, at each V_f, on an Adamel Lhomargy plastic impact tester. Fractography was conducted using a scanning electron microscope (SEM).

TABLE I Tensile properties of sunhemp fibres with standard deviations in brackets

Number of samples tested	Tensile strength (MPa)	Young's modulus (GPa)	Failure strain (%)
25	389 (±60)	35.4 (±8.8)	1.11 (±0.23)

3. Results

The tensile modulus, ultimate tensile strength and failure strain of sunhemp fibres are shown in Table I. The stress–strain curve of sunhemp fibres was found to be linear and the average failure strain was only 1.1%.

The tensile strength and modulus of composites as a function of the fibre volume per cent are shown in Figs 2 and 3, respectively. It can be seen from Figs 2 and 3 that both tensile strength and modulus increased linearly with fibre volume fraction. The solid lines in Figs 2 and 3 correspond to the computed values using the rule of mixtures (ROM) which will be discussed in the following section.

The Izod impact toughness of composites is plotted as a function of fibre volume per cent (Fig. 4). The toughness also increases linearly with fibre content as can be seen from Fig. 4 Scanning electron micrographs of the fractured surfaces (Impact) are shown in Figs 5 to 8. The average pull out length, measured using a low-power optical stereomicroscope with a graduated eyepiece was found to be 0.672 mm for 0.24 V_f composite.

4. Analysis and discussion

4.1. Tensile properties of sunhemp fibres

The high strength and modulus of the fibre can be attributed to the high cellulose content and low microfibril angle of sunhemp. Page *et al.* [4] and McLaughlin and Tait [3] have analysed the effect of microfibril angle on the behaviour of various lignocellulosics and have related this effect to the off-axis properties of short-cellulose fibre-reinforced system. The effect of cellulose content is very obvious – the

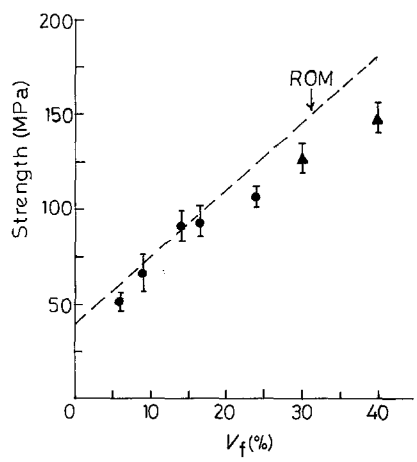

Figure 2 Tensile strength of sunhemp–polyester composites versus the fibre volume per cent. (●) Modified hand lay-up, (▲) pultruded.

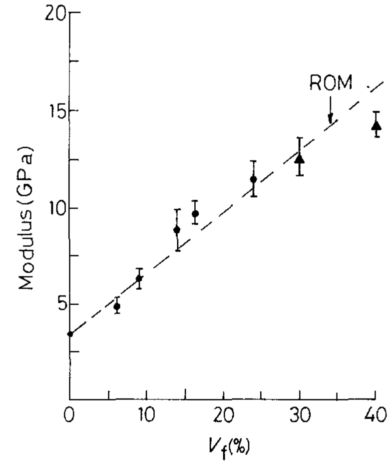

Figure 3 Young's modulus of sunhemp–polyester composites plotted against the fibre volume per cent. (●) Modified hand lay-up, (▲) pultruded.

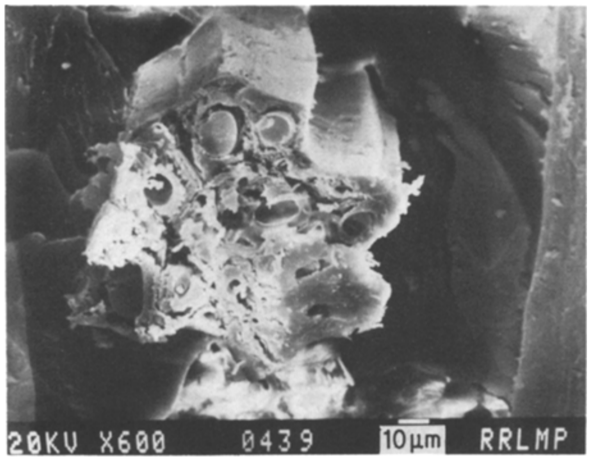

Figure 5 Fractograph of the composite showing the "corrugated" surface of the fibre and the gap created between the fibre and matrix during impact fracture.

strengthening and stiffening efficiency of natural fibres are directly related to the volume fraction of reinforcing fibres, which in this case are cellulose molecules.

4.2. Tensile behaviour of composites

The rule of mixtures (ROM) for strength of a unidirectional "continuous" fibre-reinforced material is given below:

$$\sigma_c = \sigma_f V_f + \sigma'_m (1 - V_f) \quad (1)$$

where σ_c and σ_f are the composite and ultimate fibre strength, respectively. σ'_m is the stress taken up by the matrix at the failure strain of the fibres (ε_f) and is given by $E_m \varepsilon_f$. There seems to be a good correlation between the experimental values and theory. The ROM for the tensile modulus is given by the following equation:

$$E_c = V_f E_f + (1 + V_f) E_m \quad (2)$$

where, E_c, E_f and E_m are the initial tensile moduli of the composite, fibre and matrix, respectively. The experimental behaviour of the modulus against the fibre content seems to follow the ROM quite closely. Any discrepancy may be attributed to the difficulty in computing the exact cross-sectional area on account of the irregular shape of the fibres (Fig. 5).

4.3. Fibre–matrix interfacial bond

The effectiveness of the fibre–matrix bond is dependent on the chemical compatibility and the presence of mechanical "keying" between the fibre and the matrix. The irregular surface of the fibre is likely to enhance the efficiency of the sunhemp–polyester interfacial bond, due to the greater surface area present at the interface. However, natural fibres contain waxes and fatty acid by-products [8] that are unlikely to form any chemical bonds between the fibre surface and polyester and thereby causing a weak interfacial bond. The fibre critical length, l_c, was calculated by assuming that the average pull out length, $\bar{l}_p$, is half the maximum attainable pull out length and therefore the interfacial frictional shear strength, τ_f is then obtained from the Kelly–Cottrell equation [10].

$$\tau_f = \frac{\sigma_f d}{2 l_c} \quad (3)$$

where in our case, $\tau_f = 4.34\,\text{MPa}$ and d is the fibre diameter.

4.4. Impact

There has been considerable success in modelling the toughness of glass and carbon fibre composites [11,

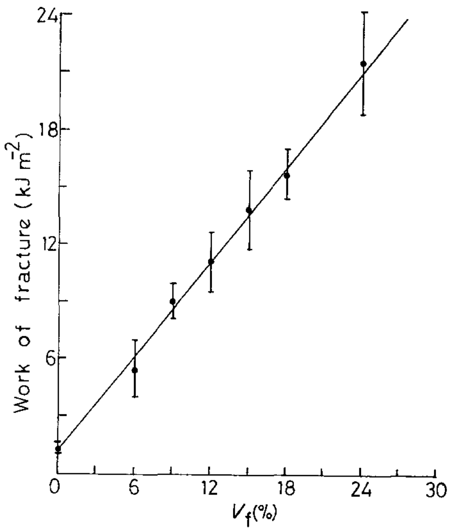

Figure 4 The work of fracture, measured by the Izod test plotted against the fibre volume per cent.

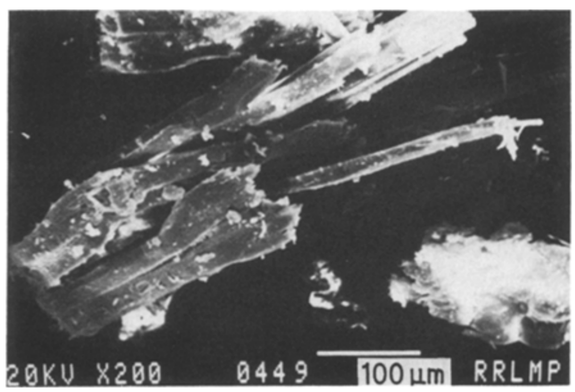

Figure 6 Fractograph of the composite showing fibre splitting and the large fibre surface area generated during impact.

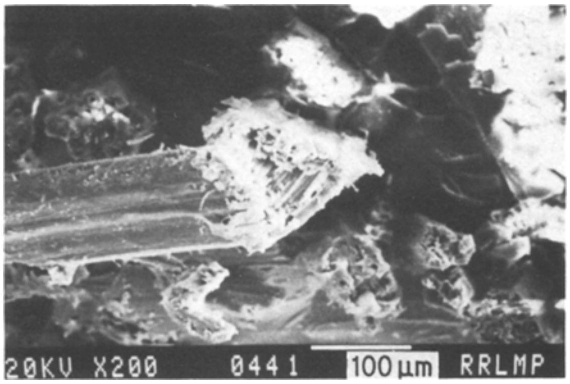

Figure 7 Fracture surface (impact) of a composite ($V_f = 0.24$) showing a pulled out fibre that has been damaged near the crack plane.

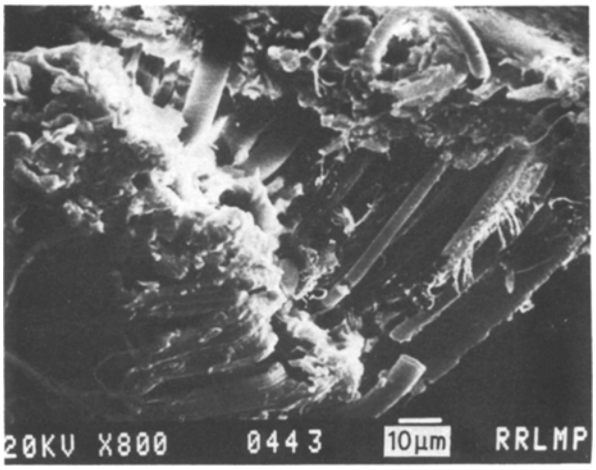

Figure 8 A magnified view of the damaged fibre (see in Fig. 7) showing the complex nature of fibre fracture.

12]. We will now try and predict the toughness of sunhemp–polyester composites using one of these models and then try and pinpoint the reasons for discrepancies, if any, between the experimental and predicted values.

Marston *et al.* [11] have predicted the toughness of fibre composites by summing the contributions of different sources of fracture mechanisms. The work of fracture (R_{total}) is considered to originate from three major fracture mechanisms and is given by [11]

$$R_{total} = R_{surface} + R_{redist} + R_{pull\ out} \qquad (4)$$

and is explained in the Appendix.

$R_{surface}$ is the contribution of fracture work by the creation of three new surfaces – fibre, matrix and fibre–matrix interface. The toughness contribution of each of these "new" surfaces will depend on the work done to create a unit area of the surface and the total area created has been analysed in detail by Marston *et al.* [11] and Harris *et al.* [12] and is given in the Appendix.

Table II shows the calculated contributions of each major energy absorbing mechanism.

It is predicted that the work to fracture the fibre–matrix interface and the fibre pull out work are the major contributions to the overall work of fracture of the composite. The predicted values overestimate the fracture work by a considerable amount.

We will now consider the two major contributions of toughness of the composite and the variations that are likely to occur by using natural fibres instead of synthetic ones.

The pull out theory assumes that the filaments break due to presence of flaws that are randomly distributed, and in the absence of this randomness the fibres will break in the crack plane and no pull out will occur. A thorough analysis of the failure of sunhemp fibre is thus needed to verify the validity of this assumption.

The frictional shear strength was calculated from the linear model (Equation 3) which assumes that the Poisson's shrinkage effect is negligible [13]. However, in the case of low modulus cellulose fibres which consist of bundles of hollow fibrils, this assumption is invalid. Secondly, when these fibres are stressed there appears to be some permanent lateral contraction that will effect τ_f. Fig. 5 shows signs of this lateral deformation – there appears to be considerable "clearance" between the fibres and the matrix, whereby no friction work is possible between the surfaces. An accurate estimate of the friction work is thus not possible unless the lateral deformation is thoroughly analysed.

The non-uniformity of the fibre will also lead to a more complex relation than used in Equation 3. Wells and Beaumont on their work on bundles of glass and carbon fibres [13] and Morrissey *et al.* [14] in sisal–cement pull out, suggest that due to the uneven fibre surface, the stressed fibre will twist and "turn" so that it can "fit" in the "socket" or "tunnel" created by the fibre (Fig. 5). This interlocking will then cause mechanical keying points between the fibre and matrix and thereby altering the frictional stress. Factors such as lateral deformation, Poissons effect and the non-uniformity of sunhemp fibres will thus effect the work of fibre pull out.

As mentioned earlier, by equating $R_{if} \simeq R_m$ an upper bound of the work of fracture of the interface (R_{if}) is utilized. R_{if} can be experimentally measured using the single fibre pull out test [15]. A value of $600\,J\,m^{-2}$ for R_{if}, also used for carbon–polyester by Marston *et al.* [11] seems more realistic than $1.365\,kJ\,m^{-2}$ which we have used. Using this value the work of fracture due to the interface decreases to $6.48\,kJ\,m^{-2}$ and the total toughness becomes $30.01\,kJ\,m^{-2}$, which is nearer to the experimental

TABLE II Toughness contribution of various sources

	$R_{surfaces}$ ($kJ\,m^{-2}$)			$R_{pull\ out}$ ($kJ\,m^{-2}$)	R_{redist} ($kJ\,m^{-2}$)	Total $R_{predicted}$ ($kJ\,m^{-2}$)	$R_{experimental}$ ($kJ\,m^{-2}$)
	Matrix	Fibre	Interface				
0.24 V_f composite	1.04	0.58	14.73	20.99	0.92	38.26	21.54

result of 21 kJ m^{-2} than the predicted value (Table II) using $R_{if} = 1.365$ kJ m^{-2}.

Another factor that needs to be considered is the complex fracture mode of sunhemp fibres as compared to the planar failure obtained for glass and carbon fibres. A great deal of fibre splitting and other energy absorbing mechanisms occur in sunhemp fibres, whereby a much larger surface area is generated, Fig. 6. Fig. 7 shows the fracture surface of a sunhemp polyester impact fracture surface showing an unusual case of a fibre which has been pulled out that is also partly damaged near the crack plane. Fig. 8 shows a magnified view of the damaged or partly fractured fibre showing fibril pull out and plastic deformation of the matrix [16].

The fibre work of fracture, R_f, was estimated from Equation A6 and σ_f and ε_f obtained at a tensile speed of 0.5 mm min^{-1}. This value may not be very realistic at the impact testing speeds which are approximately 5000 times than that of tensile testing speed. The contribution of the fibre fracture may thus be more than the predicted value of 0.58 kJ m^{-2}.

It has been assumed that the debond length, l_d, is in the range of l_c and that $l_d \simeq l_c \simeq 4\bar{l}_p$. However, Harris *et al.* [12] and Wells and Beaumont [13] have reported that debonding sometimes exceeds the critical length and this would therefore affect both the predicted energy of debonding (which is implicit in $R_{interface}$ in our case) and the energy to fracture the fibre. One would thus expect the energy to fracture the fibre will be greater than the predicted value of 0.58 kJ m^{-2}.

Toughness increased linearly with fibre content up to 0.24 V_f. However, it is not clear what will happen at higher fibre contents because of the presence of fibre–fibre interactions that may cause a change in τ_f and l_c.

5. Conclusions

Sunhemp fibres with a tensile strength of 389 MPa and having a Young's modulus (35.4 GPa) of about half that of glass fibres are potential renewable reinforcing fillers in plastics. The tensile strength of the unidirectional "continuous" sunhemp polyester composites increased linearly with V_f according to rule of mixtures. Izod impact strength also increased linearly with V_f and the impact strength of 0.24 V_f composite is 21 kJ m^{-2}. The high toughness of these natural fibre–polymer composites is attributed mainly to the fibre pull out work and the creation of a new surface at the fibre–matrix interface.

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Appendix

The total energy absorbed during the creation of three new surfaces is given by:

$$R_{surfaces} = R_m(1 - V_f) + R_f V_f + \frac{V_f l_c}{d} R_{if} \quad \text{(A1)}$$

where R_m, R_f and R_{if} are the work of fracture of the matrix, fibre and interface, respectively.

R_m, the work of fracture of the matrix, is obtained by experimentation and is 1.365 kJ m^{-2} for the polyester system used here. R_f is the energy absorbed in creating the fibre surface and is given by the equation:

$$R_f = \tfrac{1}{2}\sigma_f \varepsilon_f l_d \quad \text{(A2)}$$

where l_d is the average debonded length and is assumed to be in the order of l_c whereby $l_d \simeq l_c$.

The third surface formed by the mode I debonded ($\varepsilon_f < \varepsilon_m$) is:

$$R_{interface} = V_f \frac{l_c}{d} R_{if} \quad \text{(A3)}$$

where R_{if} is the interfacial fracture toughness. Here R_{if}, is the more difficult to determine and is approximated: $R_{if} \simeq R_m$. Since matrix material is adhering to the fibre surface, from SEM photographs of pulled out fibres, R_{if} cannot be greater than R_m and is an upper bound.

The energy absorbed by the Piggott–Fitz-Randolph stress redistribution [17, 18], which is the redistribution of energy from the fibre to the matrix after the fibre fractures. This is given by

$$R_{redist} = \frac{V_f \sigma_f^3 d}{6E_f \tau_f} \quad \text{(A4)}$$

$R_{pull\ out}$ is the work due to the friction, between the fibre and matrix, in pulling out broken fibres out of the matrix after fracture and is given by [17]:

$$R_{pull\ out} = \frac{V_f \sigma_f^2 d}{24\tau_f} \quad \text{(A5)}$$

The work of fracture would then be obtained by summing the contributions of the different sources of energy absorbing mechanisms. This is given by:

$$R = R_m(1 - V_f) + R_f V_f + \frac{V_f l_c}{d} R_m + \frac{V_f \sigma_f^3 d}{6E_f \tau_f} + \frac{V_f \sigma_f^2 d}{24\tau_f} \quad \text{(A6)}$$

Other energy absorbing mechanisms do exist, but are neglected as it is thought that in the case of the sunhemp–polyester system the other contributions are negligible.

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