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Straw-reinforced polyester composites

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Annual crop fibres are rich in cellulose and they are a cheap and rapidly renewable source of fibres with potential for polymer reinforcement. Straw fibres have been incorporated in a polyester resin matrix and the properties of the fibre and composite determined. The fibres have a Young's modulus of approximately 8 GN m^{-2} and an effective density of 5.1 kN m^{-3} when combined with resin. Useful composites can be formulated with an optimum fibre volume fraction of about 0.61, resulting in a flexural stiffness of 7.3 GN m^{-2} and flexural strength of 56 MN m^{-2}. The specific flexural stiffness is about 2.5 times greater than that of polyester resin and about half that of softwoods and GRP. The work of fracture measured in impact is about half that of softwoods. It is envisaged that alternative methods for processing the fibres and the use of a phenolic resin matrix will improve the composite properties further. Straw-based composites are suitable as core material for structural board products.

1. Introduction

Natural fibres added to synthetic polymers as fillers reduce the product cost by replacing oil-derived material with a cellulose-rich natural polymer. A further advantage is gained if natural fibre additions act as a reinforcement and improve the mechanical properties of the polymer matrix.

Timber by-products, such as wood flour, sawdust and wood chips, are commonly used in composite board products. Wood flour incorporated as a 50% filler in thermosetting phenolic resin [1] improves strength and impact resistance, and wood fibre and chips are found in many commercial products such as hardboard, medium density fibreboard and chipboard [2].

Annual crop fibres are a cheaper and more rapidly renewable source of cellulose-rich fibre [3], with a renewal time of one year as against at least thirty years for softwoods, and their full potential as a polymer reinforcement has yet to be achieved. However annual crop fibres such as jute [4, 5] and by products such as sugar cane bagasse [6] have been used as fibrous reinforcement in composites. The key to the successful performance of these fibres depends on their cellulose content and it is useful to compare the chemical composition of some plant materials (see Table I).

Wood fibres and jute contain rather more cellulose than cereal straws, but in view of the 15 to 20 million tonnes of straw burnt in EEC countries every year an investigation of straw fibres for composites applications is justified. Furthermore wheat straw offers the highest cellulose content of the common UK cereal crops. Accordingly various volume fractions of wheat straw have been combined with an unsaturated polyester resin to produce straw reinforced polyester composites. The preparation of straw fibres and their incorporation with a resin matrix are described and the resulting properties of strength, stiffness and toughness reported.

2. Structure and preparation of straw fibres

Macroscopically [8] each straw stalk is built up of sections of stem joined at nodes, which are hard bulbous areas where leaves are attached to the stem. In section, Figs. 1 and 2, the stem is seen to comprise of a dense outer layer, containing the cellulose-rich epidermis and lignin, a cellular inner region, containing the ground tissue or parenchyma, and a central void or lumen.

The parenchyma consists of vascular bundles embedded in a soft cellular material composed mainly of cellulose. The vascular bundles transport water and nutrients along the stem. The lignin material forms a thin layer around the parenchyma,

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TABLE I Composition of some plant materials after Learmonth [7]

Plant material	Cellulose (%)	Lignin (%)	Pentosans (%)
Pine	60.5	26.4	11.0
Birch	64.2	19.6	27.1
Jute	74.9	11.7	18.1
Bagasse	60.2	21.7	29.1
Wheat straw	56.7	16.6	28.4
Barley straw	48.6	16.4	31.9

Fig. 3, and although it contains vascular bundles, its main function is to stiffen the stem structure. The cellulose-rich epidermis forms a hard, rigid outer layer which protects the living cells within and stiffens the stem in conjunction with the lignin material.

The epidermis has a thin layer of silica at its outer surface [9] which inhibits good bond formation with polyester resins [10]. Caustic ammonia or soda will remove the silica rich layer and improve surface adhesion. In order to avoid an extra chemical processing step in fibre preparation the straw was mechanically crushed by rolling to improve the fibre to resin bond. In addition this treatment reduced the porosity, and hence the air content, of the parenchyma and lumen, and sheared the hard epidermis allowing easy penetration of the resin to the softer tissue beneath. A polyester matrix was selected because of its successful use in jute–polyester composites [4] and its lower shrinkage compared with the more cellulose-compatible phenolic resins.

A straw stem, lightly compressed in a hand press at 1 MN m^{-2} is viewed in Fig. 4 and sketched diagramatically in Fig. 5. At this stage the porosity is still substantial and the plane of weakness introduced between the inner layers of the fibres results

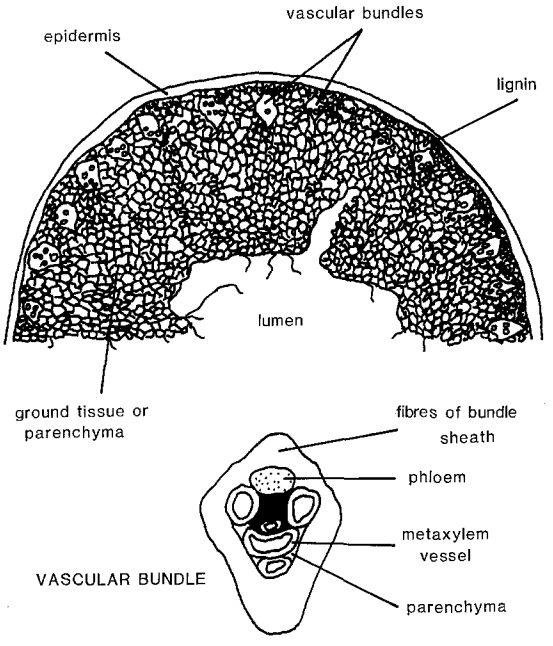

Figure 2 Cross-section of stem and vascular bundle.

in an incoherent composite product when combined with the resin matrix. To allow greater resin penetration to the inner porous layers, thereby improving resin adhesion, the approximately 4 mm diameter straws were rolled flat in a mill with a 0.25 mm clearance between the rollers. The fibres were split longitudinally and deformed in shear, rupturing the hard epidermis, Fig. 6. Fibre to resin adhesion was significantly improved and porosity reduced in the composite.

3. Properties of straw fibres

3.1. Stiffness and strength

Straight, flaw-free lengths of uncrushed and rolled fibres were selected and values of the fibre

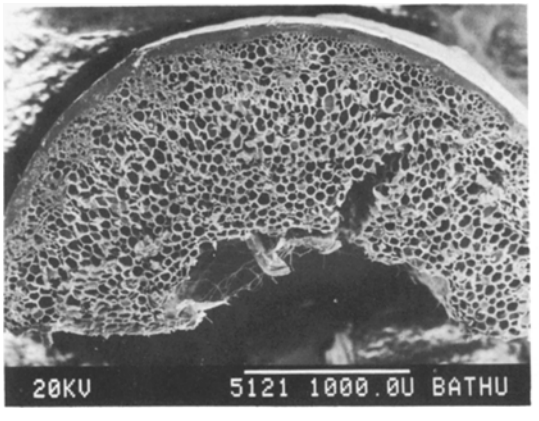

Figure 1 Cross-section of wheat straw stem.

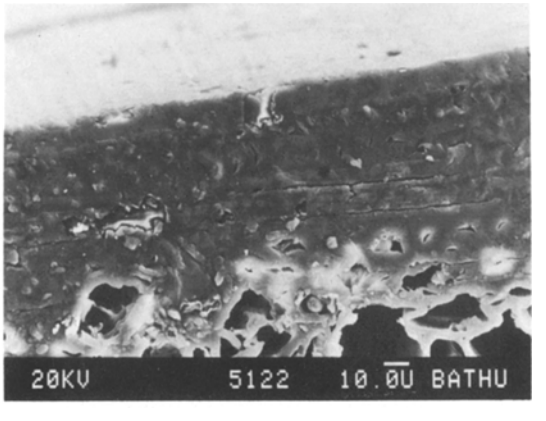

Figure 3 Epidermis and lignin layers.

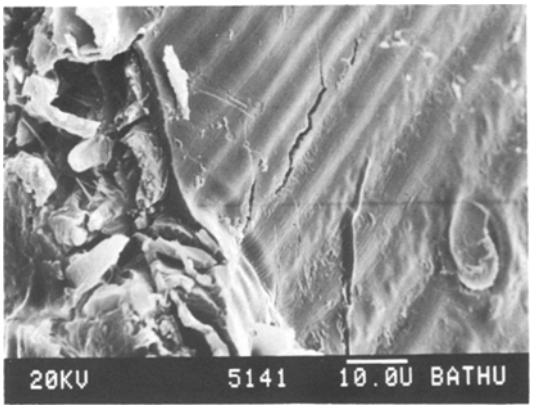

Figure 4 Pressed straw stem with cracked epidermis.

Young's modulus and strength were determined. Uncrushed fibres were held in the Instron testing machine grips via wooden dowels inserted into the fibre ends and glued with epoxy resin. Crushed fibres were gripped via aluminium tabs glued to the fibre ends. Tests were performed at a cross-head speed of $0.2\,\text{mm}\,\text{min}^{-1}$ and strain was measured with an extensometer. In the calculation of stress the cross-sectional area of the uncrushed fibre was taken as the area of the non-porous outer lignin and epidermis layers and that of the crushed fibres as the total cross-sectional area. Clearly some error must result due to variation in the wall thickness of natural fibres. The average Young's modulus and strength of uncrushed and crushed fibres derived from the close to linear stress–strain characteristics are summarized in Table II.

Scatter in the results for the compressed fibres is rather less than for the unmodified fibres due probably to a more accurate determination of the cross-sectional area and a more consistent failure mode. Fibre fracture was discontinuous with evidence of a fine fibrous microstructure and pull-out of cellulose microfibrils in the failed straw stem walls.

3.2. Fibre volume fraction and density

The density and volume fraction of crushed fibres incorporated in a cured polyester resin matrix was calculated by two methods to enable rule of mixtures analyses of measured composite properties to be made.

Firstly a known weight of fibres was combined with a known volume of uncured resin and the mixture was degassed in a vacuum desiccator. The resin was cured and the volume change Δv_1 after curing compared with the volume change Δv_2 of an equal volume of resin cured alone. The fibre volume, $(\Delta v_1 - \Delta v_2)$, and thus the volume fraction could then be calculated. The effective fibre wall density in a cured resin matrix, ignoring the volume of the resin-filled lumen and surface cracks, could also be simply calculated knowing the original fibre mass.

The second method involved measuring the density, ρ_c, of the composite of mass M_c at a given mass fraction of resin M_R. Volume fraction of resin,

$$V_R = \frac{M_R}{M_c}\frac{\rho_c}{\rho_R} \quad (1)$$

where ρ_R is the resin density measured to be $11.28\,\text{kN}\,\text{m}^{-3}$. Then the volume fraction of straw,

$$V_F = 1 - V_R \quad (2)$$

and the fibre density

$$\rho_F = \frac{M_F}{M_c}\frac{\rho_c}{V_F}. \quad (3)$$

The two methods produced similar results and an average value for fibre density of $5.1\,\text{kN}\,\text{m}^{-3}$ was assumed in all calculations.

4. Fabrication of composites and test methods

4.1. Fabrication

Rolled, flat fibres of various lengths, ranging from small fragments to several centrimetre lengths, were accurately weighed and combined with unsaturated Strand Glass polyester Resin A and catalyst in an open-ended mould with a close fitting plunger. Layers of fibre were placed in the mould, alternating with layers of resin until all the fibres were used, starting and ending with

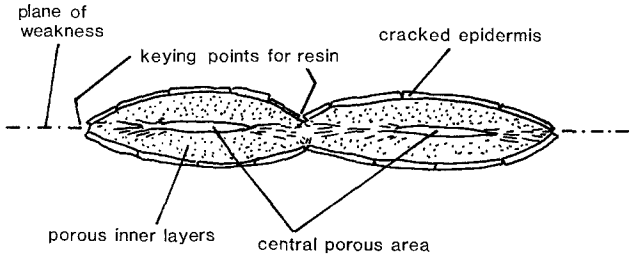

Figure 5 Cross-section of partially crushed straw fibre.

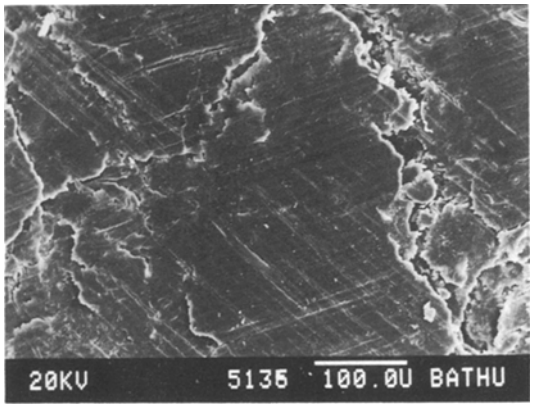

Figure 6 Ruptured epidermis after rolling.

layers of resin. A maximum of 15 min was available for laying up before the onset of air cure.

The plunger was immediately fitted and pressed onto the composite at a pressure of 5 MN m^{-2} for up to 18 h. The composites were post-cured for 2 h at 80° C before removal from the mould. Weighing of the samples enabled the weight percentage of fibres to be calculated and hence the volume fraction from Equations 1 and 2.

In addition to the samples with randomly selected fibres a composite using selected fibres not shorter than 6 cm in length was made using the above technique with fibres aligned in parallel at 40 wt% fibre. The maximum weight fraction of fibre was limited to 50% as composites with higher weight fractions could not be removed from the mould without sample delamination. Moulded samples were 300 mm in length, 15 mm wide and 3 mm thick and smaller samples were cut for testing according to the test geometry required.

4.2. Flexural and tensile tests

Three point bend tests were performed in accordance with ASTM D790, method 1, procedure A to measure flexural stiffness and strength. The samples were 75 mm long by 15 mm wide by 3 mm thick and eight tests were carried out for each composition. The outer rollers were 50 mm apart and the samples were tested at a strain rate of 0.5 mm min^{-1}. A three point bend test was chosen in preference to a four point bend test because although it underestimates the flexural modulus it requires less material for each test and eliminates the need to accurately determine centre point deflections with a transducer.

The flexural modulus,

$$E_B = \frac{L^3 m}{4bd^3} \quad (4)$$

and the maximum fibre stress,

$$S = \frac{3PL}{2bd^2} \quad (5)$$

where L is the support span (50 mm), b is the width, d is the thickness, P is the maximum load, and m is the slope of the initial straight line portion of the load-deflection curve.

Tensile test pieces were un-necked with the same width and thickness of the flexural specimens. Overlapping aluminium tabs were glued to the ends of the specimen with epoxy resin filling the space at the tab overlap to prevent compression of the sample at the grip. The samples were tested at a cross-head speed of 0.5 mm min^{-1} and strain measured with an extensometer.

4.3. Impact testing

Test pieces were 40 mm long, 8 mm wide and 3 mm thick with a 1 mm deep saw cut at the centre of the edge face. A sharp knife blade was drawn across the centre of the saw cut at 90° to the sample axis to obtain a consistent starter crack. The samples were fractured in a Hounsfield plastics impact machine and the impact toughness calculated from the energy absorbed and the sample cross-sectional area behind the notch.

5. Results and analysis

5.1. Flexural tests

A typical load–deflection curve for a 40 wt% (volume fraction = 0.61) composite is presented in Fig. 7. Arrest points correspond to breakage and pull-out of individual fibres from the resin matrix. The flexural modulus was calculated from the initial close to linear portion of the curve and the

TABLE II Young's modulus and strength of uncrushed and crushed straw fibres, with standard deviations in brackets

Fibre type	Number of samples	Young's modulus (GN m^{-2})	Strength (MN m^{-2})
Uncrushed	20	9.3 (1.7)	43.1 (5.6)
Crushed	20	8.1 (0.35)	36.2 (3.1)

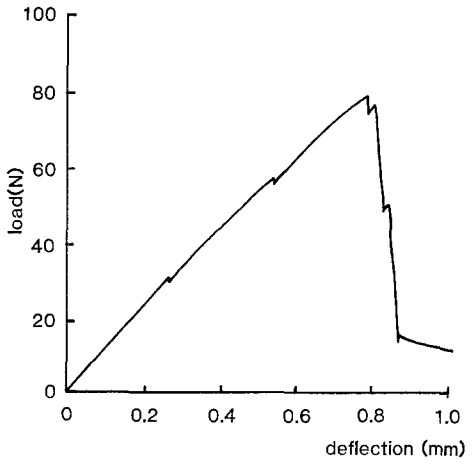

Figure 7 Load against deflection characteristic for $V_F = 0.61$ straw composite tested in flexure to failure.

maximum fibre stress at the point of maximum load. At low fibre volume fractions the load–deflection curves are smooth to failure suggesting that the straw fibres act more as impurities than reinforcement.

Results for all volume fractions of straw fibre are included in Table III with standard deviations in parentheses. Tensile modulus determinations are included here for comparison. As the fibre volume fraction increases from zero the initial stiffness and strength decreases ($V_F = 0.21$) but the properties steadily improve to a maximum in strength at $V_F = 0.61$ and in Young's modulus at $V_F = 0.7$. The samples with carefully aligned fibres at $V_F = 0.61$ have significantly improved properties compared with the other samples containing random lengths of short misaligned fibres. The reduction in the composite density with increasing fibre content has a beneficial effect on the composite specific stiffness. The 0.61 volume fraction composite has a flexural stiffness of 0.99 ($m \times 10^6$) a factor of 2.5 times greater than the pure resin, indicating significant commercial promise. In comparison softwoods and GRP (glass reinforced polymers) have specific stiffnesses of approximately 2.0, high tensile steel, 2.7, and CFRP (carbon fibre-reinforced plastic) ($V_F = 0.6$), 8.7.

The flexural strength of the composites is plotted against fibre volume fraction in Fig. 8. The results can be fitted to two theoretical linear curves which help to explain the development of the strength in the composites. Assuming that the elastic strains in the composite, matrix and fibre are all equal, at low fibre volume fractions the strength of the composite, σ_c, is governed by an equation of the form,

$$\sigma_c = \sigma_R(1 - V_F) \quad (6)$$

where σ_R = strength of the resin and V_F = volume fraction of the fibre.

The matrix failure strain is assumed to be higher than the fibre failure strain. As the volume fraction of fibre increases a modified rule of mixtures expression predicts,

$$\sigma_c = \sigma_F V_F + \sigma'_R(1 - V_F) \quad (7)$$

where σ_F = fibre strength and σ'_R = stress in resin at fibre breaking strain.

At a fibre volume fraction V_{min} where the composite strength is a minimum,

$$\sigma_F V_{min} + \sigma'_R(1 - V_{min}) = \sigma_R(1 - V_{min}) \quad (8)$$

TABLE III Mean flexural and tensile properties of straw composites as a function of fibre content

Weight fibre (%)	Volume fibre (%)	Density ρ_c (kN m^{-3})	Flexural			Tensile	
			E (GN m^{-2})	σ_{max} (MN m^{-2})	E/ρ ($m \times 10^6$)	E (GN m^{-2})	E/ρ ($m \times 10^6$)
0	0	11.28	4.4 (± 0.05)	32 (± 0.05)	0.39	3.3 (± 0.30)	0.29
10	21	9.91 (± 0.68)	2.7 (± 0.10)	25 (± 0.3)	0.27	4.2 (± 0.44)	0.42
20	37	8.83 (± 0.68)	4.7 (± 0.39)	40 (± 5.6)	0.53	4.7 (± 0.42)	0.53
30	50	8.04 (± 0.49)	4.9 (± 0.40)	47 (± 6.0)	0.61	5.6 (± 0.45)	0.70
40	61	7.36 (± 0.39)	5.5 (± 0.45)	53 (± 5.2)	0.75	8.2 (± 0.35)	1.11
40*	61	7.36 (± 0.39)	7.3 (± 0.40)	56 (± 4.3)	0.99	8.0 (± 0.30)	1.09
50	70	6.77 (± 0.29)	6.2 (± 0.35)	34 (± 2.5)	0.92	–	–

40* contains 40 wt% of selected aligned fibres. The standard deviation is recorded in parentheses.

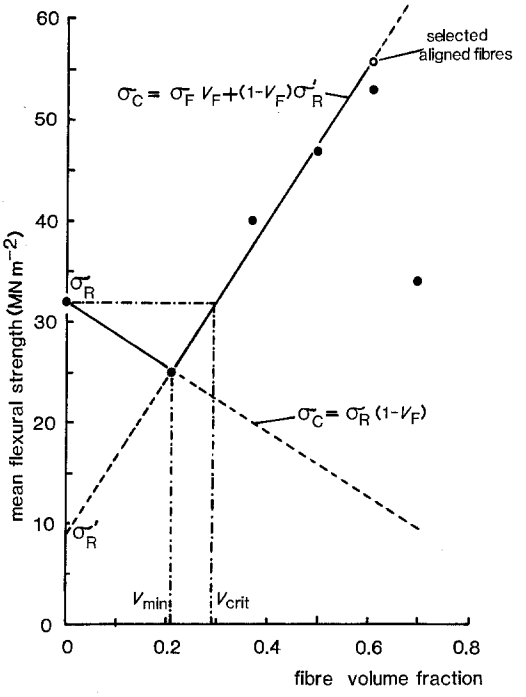

Figure 8 Mean flexural strength of straw composite against volume fraction of fibre, including theoretical curves for composite strength.

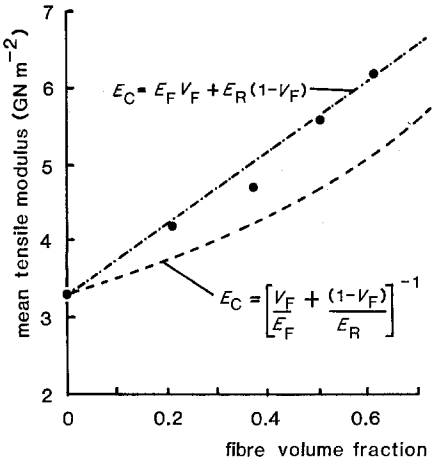

Figure 9 Mean tensile Young's modulus of composite against volume fraction of straw fibre, including theoretical curves for composite Young's modulus.

from Equations 6 and 7 and rearranging terms,

$$V_{min} = \frac{(\sigma_{R'} - \sigma_R)}{(\sigma_{R'} - \sigma_R - \sigma_F)} \quad (9)$$

Values for $\sigma_{R'}$ and σ_F may be obtained by extrapolating the experimental line which passes through the σ_c values at $V_F \geqslant 0.21$ (Fig. 8) in both directions. A value for the fibre strength of 85.9 MN m^{-2} is obtained approximately double the experimentally determined values for single fibres. The single fibre results are undoubtedly depressed due to their sensitivity to surface flaws. The stress in the resin at the fibre breaking strain $\sigma_{R'}$ is graphically determined to be 9.0 MN m^{-2}. By calculation V_{min} equals 0.21.

The second critical volume fraction, V_{crit}, is defined as the fibre volume fraction where the composite strength is equal to the strength of the pure matrix and represents the minimum volume fraction for a useful composite. At this point,

$$\sigma_R = \sigma_F V_F + \sigma_{R'}(1 - V_F) \quad (10)$$

and $V_{crit} = (\sigma_R - \sigma_{R'})/(\sigma_F - \sigma_{R'})$ resulting in a calculated value of $V_{crit} = 0.29$.

Although the above treatment fits the limited experimental data well, it does not take into account the loss of strength above $V_F = 0.61$. Here it is proposed that resin depletion between fibres results in a poor inter-fibre bond allowing the fibres to shear past each other in the three-point bend test, introducing planes of weakness parallel to the fibres.

A fall in the composite flexural modulus occurs at $V_F = 0.21$. It is thought that in-plane shear stresses and compression stresses, which may possibly buckle fibres, account for this unexpected initial fall in modulus.

5.2. Tensile tests

In-plane shear stresses and compression stresses acting on straw composite samples tested in three-point bending are absent in material tested in pure tension. As a result there is no initial fall in the value of the tensile modulus, Table III and Fig. 9, as the fibre volume fraction increases. A rule of mixtures relationship for tensile modulus, similar in form to Equation 7,

$$E_c = E_F V_F + E_R(1 - V_F) \quad (12)$$

is unlikely to predict the composite modulus due to the random orientation and variation in the length of the straw fibres. The equation represents an upper limit of the modulus which cannot be exceeded and assumes that the fibre and matrix undergo the same strain and are stressed along a unidirectional fibre axis. A lower limit of modulus is predicted by,

$$E_c = \left[\frac{V_F}{E_F} + \left(\frac{1 - V_F}{E_R}\right)\right]^{-1} \quad (13)$$

which assumes that the composite is stressed

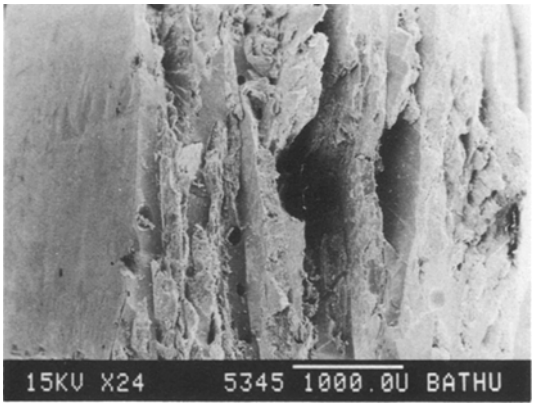

Figure 10 Tensile fracture surface of $V_F = 0.61$ composite.

perpendicular to the straw fibre axis and the stresses in the fibre and matrix are the same. The tensile modulus results, Fig. 9, are seen to fall between the two extremes of Equations 12 and 13 which might be expected from the orientation of fibres. Long fibres are oriented closely parallel to the specimen axis whereas short fibres are more randomly oriented.

The fracture topographies of straw composite failed in tension and three-point bending may account for the differences between the tensile and flexural modulus at low fibre volume fractions. A tensile failure surface ($V_F = 0.61$), Fig. 10, shows evidence of brittle resin and fibre failure and short fibre pull-out. The resin is in intimate contact with the rectangular section crushed straw fibres and resin penetration is good. Resin surfaces, revealed after pull-out, bear the imprint of the hard straw epidermis, and confirm that the resin to fibre bond is weak due to the silica-rich layer. Fibre fragments indicate that fibre fracture occurs at the porous inner layer between the hard epidermis layers.

Tensile failures are relatively flat and transverse. Bend failure surfaces are considerably greater in area and indicate that extensive shear delamination has occurred at the fibre to resin interface and within the parenchyma tissue. The action of in-plane shear and compressive stresses in the three-point bend tests appears to account for the initial fall in modulus at low fibre volume fractions.

5.3. Impact tests

The results of pendulum impact tests, Fig. 11, which are effectively high strain rate three-point bend tests reflect the trends in change in the flexural modulus and strength with fibre volume

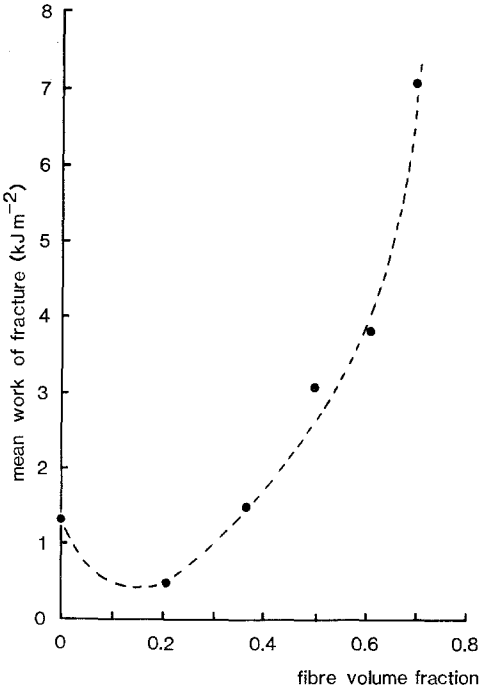

Figure 11 Mean work of fracture against fibre volume fraction for straw composite tested in impact.

fraction. A minimum composite toughness occurs at about $V_F = 0.21$. At $V_F = 0.70$ the composite has a work of fracture close to 10 kJ m^{-2}, which almost places the straw composite in the category of tough engineering materials, (> 10 kJ m^{-2}). Softwoods have toughness values of approximately 20 kJ m^{-2}.

The crushed straw fibres are a mixture of platelets and longer crushed fibres of approximately rectangular cross-section. In impact the straw fibres are capable of arresting and diverting the progress of propagating cracks from an edge notch, thus blunting the crack by delamination.

6. Conclusions

The foregoing results have demonstrated that a useful composite material can be manufactured from straw and polyester resin. The straw fibres considerably improve the stiffness, strength and toughness of the resin alone, and reduce its density. As well as the straw acting as a cost-reducing agent the composite can be regarded as a successful engineering material in its own right, particularly as a lightweight building material.

In order to exploit straw as a reinforcement, for example, in constructional boards it is important that the cost of processing the raw fibre, prior

to incorporation with resin, is minimized. Future work will investigate the performance of other lower cost resin systems, particularly phenolic resins, and alternative pre-treatments of straw fibres. The cost of chemically removing the silica-rich layer at the surface of the epidermis or plasticizing the fibre may outweigh any improvements in the mechanical performance, however. Straw composite properties can be significantly improved by the selection and alignment of long fibres and it is by careful handling and processing of fibres during composite manufacture that optimum properties will be achieved.

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