

# Effects of water and bleaching on the mechanical properties of cellulose fibre cements\*

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The mechanical properties of bleached and unbleached cellulose fibre-cement composites are studied in both dry and wet conditions. Bleaching the fibres increases both the elastic modulus and flexural strength but reduces the specific work of fracture. Water has a dramatic adverse effect on the elastic modulus and flexural strength of the composites irrespective of whether the fibres are bleached or not. However, water increases remarkably the fracture resistance. A failure mechanism is proposed to explain these experimental results and this is supported by evidence obtained from scanning electron micrographs of fracture surfaces.

## 1. Introduction

Cellulose fibres have been considered serious candidates for replacing asbestos in cement matrices. This is because of their low cost and reasonably good mechanical properties. We have shown in a previous paper [1] that the cellulose fibre-reinforced composite possesses sufficient strength and fracture resistance to render it useful for many non-structural applications. These properties may be further improved if stronger cellulose fibres are used and if the fibre-matrix bond can be enhanced by chemical treatment with certain types of coupling agents [2]. The Hatschek process is used in Australia to produce cement sheets reinforced with cellulose fibres. This involves an autoclave process to obtain optimum properties of the cement matrices. Fibres derived from the kraft pulp contain a small amount of lignin, and it has been suggested that during autoclaving the lignin compounds formed can cause an inhibiting effect on the curing of the cement [3]. To overcome this problem the fibres can be subjected to a bleaching process to remove the lignin. This paper investigates the effects of bleaching cellulose fibres of the Wisakraft source on the mechanical properties of cellulose-cement composites. The experimental results are discussed and compared with those

obtained from control cement sheets reinforced with Kinleith fibres. Tests were conducted in both dry and wet conditions. The mechanism of failure is also examined with the aid of a scanning electron microscope (SEM).

## 2. Experimental details

### 2.1. Material

The cellulose-cement composites were prepared in the form of 5 mm thick sheets in the Engineering and Research Laboratories of James Hardie & Coy Pty. Ltd by the Hatschek process. These sheets contained cement/silica in the ratio 50:50 and had a fibre mass fraction of about 8%. After curing for 24 h at 100% relative humidity they were autoclaved at 140°C and 0.4 MPa for 24 h. The densities of the resultant composites were in the range of 1.23 to 1.30 g cm<sup>-3</sup>. The cellulose fibres used in this work were supplied from the following sources: 1. test pulps: unbleached and bleached pulps were obtained from "Schanman Ab. Oywilh", Helsinki Mills, Jakobstad, Finland; and 2. control pulp: New Zealand Forest Products, unbleached Kinleith kraft.

### 2.2. Testing methods

Rectangular samples measuring 150 mm × 24 mm

\*An earlier version of this paper was presented at the Eighth Australian Conference on the Mechanics of Structures and Materials, University of Newcastle, NSW, Australia, August 1982.

TABLE I Mechanical properties of bleached Wisakraft cellulose fibre–cement composites

Test condition	Young's modulus, $E_b$ (GPa)		Flexural strength, $\sigma_b$ (MPa)		Fracture toughness, $R$ ( $\text{kJ m}^{-2}$ )	
	MD	CD	MD	CD	MD	CD
As-received	10.38 $\pm$ 1.24	9.07 $\pm$ 0.84	22.4 $\pm$ 1.66	17.31 $\pm$ 1.17	0.34 $\pm$ 0.031	0.26 $\pm$ 0.016
oven dried						
Immersed in water	7.0 $\pm$ 1.06	3.98 $\pm$ 0.62	18.92 $\pm$ 1.48	13.25 $\pm$ 0.96	2.51 $\pm$ 0.35	2.3 $\pm$ 0.22
Oven dried after immersion in water	9.02 $\pm$ 0.38	8.90 $\pm$ 0.56	23.2 $\pm$ 0.97	20.42 $\pm$ 0.76	0.37 $\pm$ 0.044	0.34 $\pm$ 0.024

N.B. MD = machine direction; CD = cross direction.

$\times 5$  mm were cut from the cement sheets in both the machine and cross directions (MD and CD) for three-point loading flexural and work of fracture tests. All experiments were performed in the Instron testing machine at a crosshead speed of  $2 \text{ mm min}^{-1}$ . The Young's modulus in bending ( $E_b$ ) was calculated from the slope of the load–displacement curve and the flexural strength ( $\sigma_b$ ) was determined by the maximum load-to-failure. For the specific work of fracture ( $R$ ) measurements, a notch of depth 12 mm was introduced at the mid-section of the specimen and the total area under the load–displacement curve was used to obtain  $R$  by dividing it by the fracture ligament area. All tests were performed on wet as well as dry samples. Dry samples were oven dried at a temperature of about  $120^\circ \text{C}$  for approximately 24 h before testing, while wet samples were prepared by immersion in water for 72 h prior to testing. Tests were also conducted on samples previously wet and then dried in the oven.

### 3. Results and discussion

#### 3.1. Strength and modulus results

The modulus and strength properties tested at different conditions are given in Tables I, II and

III for the two test pulps and the control pulp, respectively. Two general observations can be made from these results.

##### 3.1.1. Orientation effect

From Tables I and II it appears that the modulus (in wet conditions) and strength (both wet and dry) of the composites with the Wisakraft fibres are direction dependent, with  $E_b$  and  $\sigma_b$  values in the MD slightly higher than those in the CD. This orientation effect is more evident in the Kinleith cellulose–cement composite, Table III. An investigation into the fibre orientation was carried out on these cement composite sheets. The technique used was to polish a few samples and then etch in hydrochloric acid to expose the fibres. The samples were then prepared for SEM examination. Fig. 1 clearly demonstrates a specific alignment of the fibres along the length of the sheet. This preferential orientation must therefore be the reason for the higher flexural strength and larger elastic modulus ( $E_b$ ) in the machine direction.

##### 3.1.2. Water effect

In Tables I, II and III, the flexural strength ( $\sigma_b$ ) values for the Wisakraft (bleached and unbleached)

TABLE II Mechanical properties of unbleached Wisakraft cellulose fibre–cement composites

Test condition	Young's modulus, $E_b$ (GPa)		Modulus of rupture, $\sigma_b$ (MPa)		Fracture toughness, $R$ ( $\text{kJ m}^{-2}$ )	
	MD	CD	MD	CD	MD	CD
As-received	7.97 $\pm$ 1.26	7.64 $\pm$ 1.10	19.23 $\pm$ 1.64	15.43 $\pm$ 1.68	0.48 $\pm$ 0.03	0.36 $\pm$ 0.035
oven dried						
Immersed in water	5.58 $\pm$ 1.24	3.78 $\pm$ 1.16	14.88 $\pm$ 2.20	11.66 $\pm$ 1.6	2.69 $\pm$ 0.12	2.30 $\pm$ 0.14
Oven dried after immersion in water	8.1 $\pm$ 0.61	6.93 $\pm$ 0.81	20.07 $\pm$ 1.56	15.27 $\pm$ 1.30	0.46 $\pm$ 0.03	0.37 $\pm$ 0.014

N.B. MD = machine direction; CD = cross direction.

TABLE III Mechanical properties of unbleached Kinleith kraft cellulose fibre–cement mortar composites

Test condition	Young's modulus, $E_b$ (GPa)		Flexural strength, $\sigma_b$ (MPa)		Fracture toughness, $R$ ( $\text{kJ m}^{-2}$ )	
	MD	CD	MD	CD	MD	CD
As-received	$4.5 \pm 1.42$	$3.46 \pm 0.43$	$18.96 \pm 0.59$	$8.62 \pm 0.97$	—	—
oven dried						
Immersed in water	$3.46 \pm 0.56$	$2.25 \pm 0.58$	$10.70 \pm 1.15$	$3.96 \pm 0.72$	$4.41 \pm 0.63$	$1.73 \pm 0.14$
Oven dried after immersion in water						
	$5.15 \pm 1.10$	$3.28 \pm 0.53$	$16.99 \pm 1.61$	$8.70 \pm 1.17$	$1.32 \pm 0.074$	$0.32 \pm 0.073$

N.B. MD = machine direction; CD = cross direction.

and the control Kinleith cement composites are much lower for wet samples, indicating a weakening effect due to the presence of water. However, the removal of water from the material by the process of redrying brings it back to its original strength. A statistical  $t$  test has given only a slight difference in  $\sigma_b$  which confirms that the effect of water on the cellulose cement composites is reversible. A similar behaviour is also obtained for the Young's modulus (see Tables I, II and III).

### 3.2. Fracture toughness results

For fracture toughness tests, Figs. 2 and 3 show some typical load–displacement curves for the notched beams under three-point loading. Clearly, crack propagation in all the composites is more stable in the wet state.  $R$  values obtained in these experiments are given in Tables I to III and they indicate a similar trend to that of  $\sigma_b$  and  $E_b$ , i.e. larger  $R$  values in the MD and a considerable increase for those samples soaked in water are obtained.



Figure 1 Kinleith cellulose fibres in cement matrix. Alignment of fibres is along the machine direction (120 X).

### 3.3. Bleached versus unbleached Wisakraft cellulose fibre–cement composites

A comparison of the results given in Tables I and II shows that the bleached Wisakraft cellulose fibre cements have larger stiffness ( $E_b$ ) and strength ( $\sigma_b$ ), in both directions, than the unbleached Wisakraft cellulose fibre cements. The failure behaviours in these two types of cellulose fibre cements with respect to the load–displacement curves are different. In bending tests, fracture is more stable and ductile in the unbleached cellulose fibre cements; but in the bleached cellulose fibre cements failure is more unstable and brittle with a precipitous load drop after the maximum is reached. Typical load–displacement records are given in Fig. 4 to illustrate the different failure behaviours. With respect to the fracture toughness, higher  $R$  values are obtained for the unbleached fibre–cements in both directions (refer to Tables I and II). In the presence of water, the performances of the two types of cellulose composites change with a drop in stiffness and strength, and an increase in toughness.

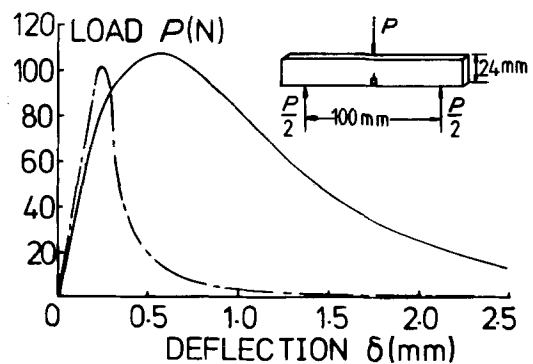


Figure 2 Specific work of fracture tests under three-point bending. Unbleached fibre–cement composite. (— for wet sample; --- for dry sample.)

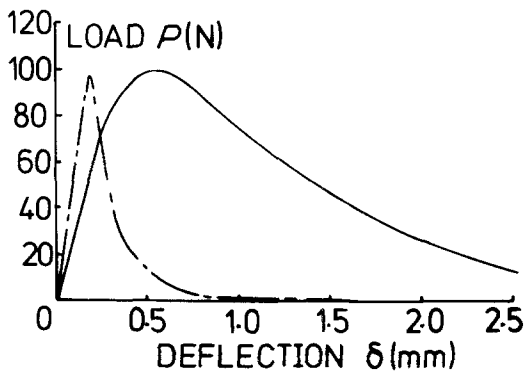


Figure 3 Specific work of fracture tests under three-point bending. Bleached fibre-cement composite. (— for wet sample; --- for dry sample.)

### 3.4. Wisakraft versus Kinleith kraft fibre cement composites

It may be inferred from the mechanical properties shown in Tables I to III that the Wisakraft fibre composites have much better strength and elastic modulus but poorer fracture toughness than the Kinleith kraft fibre composites. Both Wisakraft and Kinleith kraft fibre cements have weaker strengths and higher toughnesses in wet conditions. When these properties are compared with those obtained in dry conditions the Wisakraft fibre composites show less reduction in strength but greater improvement in toughness than the control Kinleith kraft fibre composites for both the machine and cross directions. The wet strength properties may be improved if water can be prevented from getting to the cellulose fibres. A plausible technique is to pre-coat the fibres with resins before mixing with the cement slurry.

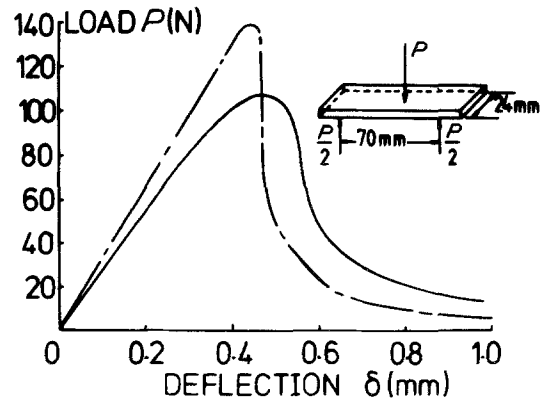


Figure 4 Typical load-displacement curves for the three-point bending test. — for unbleached fibre-cement composite; --- for bleached fibre-cement composite.

Development and research work are in progress in this direction. Previous work has also shown that epoxy resin improves the properties of the cement matrices.

### 4. SEM study on fracture surfaces

An important feature which is evident from the experimental results is the effect of water on these cellulose-cement composites. This is with respect to the reduction in strength and the increase in toughness. The understanding of such a phenomenon requires the aid of an SEM study, which was carried out on the fracture surfaces produced from the control Kinleith pulp. Typical fracture surfaces in both dry and wet environments are shown in Figs. 5 and 6. The overall picture clearly shows an apparently more dense cellulose fibre concentration on the surface in the wet condition for both

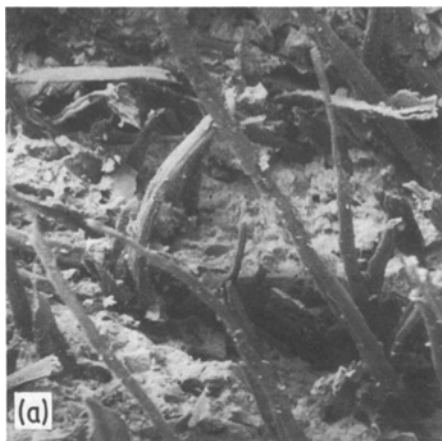


Figure 5 Fracture surfaces of oven dried Kinleith cellulose-cement composite (120 $\times$ ). (a) Machine direction; and (b) cross direction.

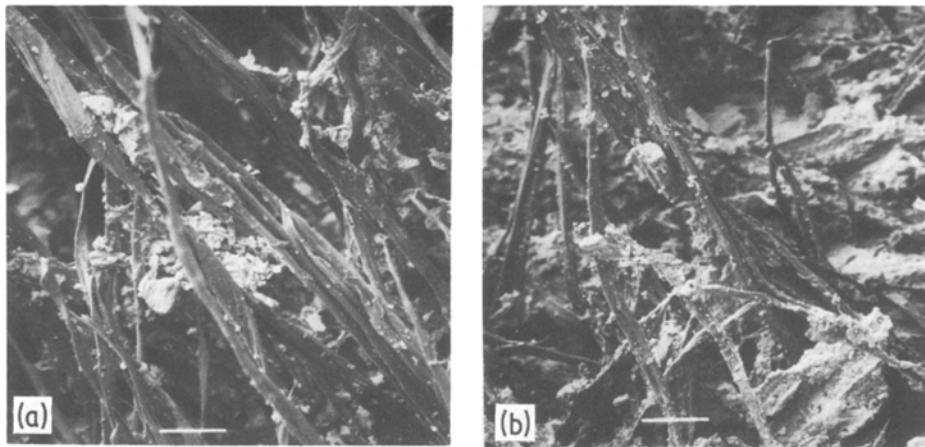


Figure 6 Fracture surfaces of Kinleith cellulose–cement composite tested in wet conditions (120 ×). (a) Machine direction; and (b) cross direction.

machine and cross directions. A close examination was carried out on the tips of the cellulose fibres. In the dry condition, Fig. 7, failure in many fibres was brittle with little or no reduction in cross-sectional area and these fibres remain close to the fracture surface. There are relatively few unbroken fibres. In the wet condition, Fig. 8, the fibres showed a reduction in cross-sectional area and appeared to have untwisted and stretched during pull-out, thus giving rise to the presence of many long fibres on the fracture surface. Broken fibres are rare in this case. We propose therefore that the mechanism of failure in these cement composites in the wet and dry conditions is controlled mainly by the behaviour and physical properties of the cellulose fibres. Dry cellulose fibres are strong and brittle and have a reasonable fibre–matrix bond to enable fibre fracture to occur. Thus, the strength

of the composite is high but the toughness value is low because of the small pull-out length of the fibres. Wet cellulose fibres, on the other hand, are ductile in behaviour due to the stretching and unravelling of the fibres to give large pull-out fibre lengths. Consequently, there are increases in the fracture toughness but reductions in the flexural strength of the wet composites. This toughening mechanism is similar to that observed for wood where single wood tracheids under tensile load can either buckle [5] or untwist [6] depending on end conditions. These behaviours are caused by the helical pattern of the cellulose microfibrils in the secondary  $S_2$  wall and they both give rise to large toughnesses in wood [7]. It is not clear how effectively water can affect the fibre–matrix interfacial bond strength, since fibre swelling will increase the frictional strength, but as fibre pull-

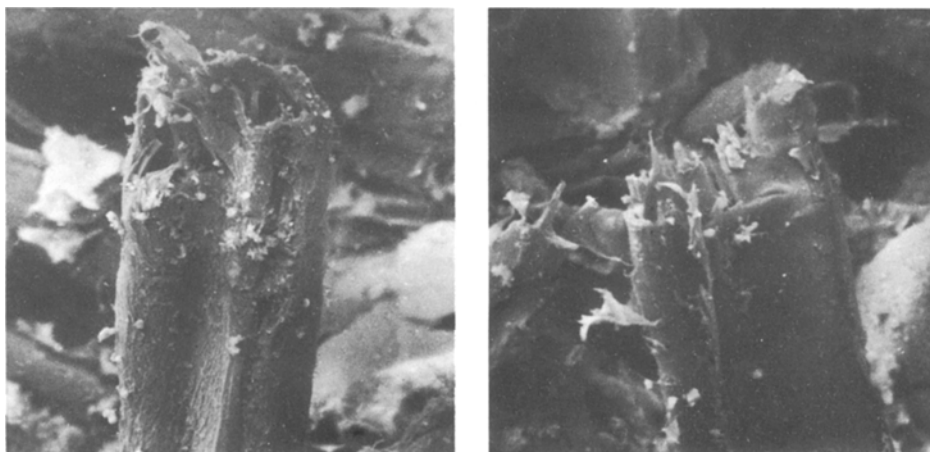


Figure 7 Kinleith cellulose fibres: brittle type failure (750 ×).

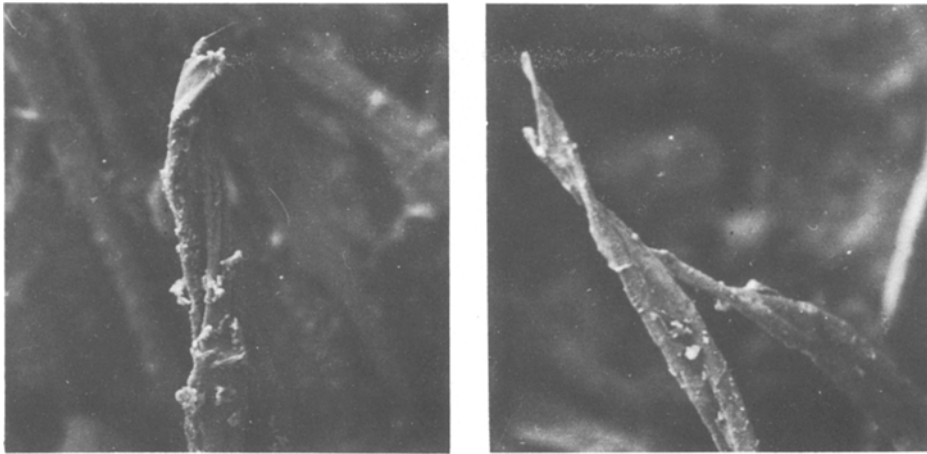


Figure 8 Kinleith cellulose fibres: ductile failure (340 ×).

out occurs the continuous fibre unravelling will reduce it.

Further SEM micrographs to support the above proposed failure mechanism are shown in Figs. 9 and 10. These are the fracture surfaces of samples tested in the flat-wise direction across their widths. In dry conditions, again most of the fibres are broken and protruding on each side of the fractured specimen, Fig. 9. In the wet condition, the fibres, due to their enhanced ductility caused by stretching and unravelling in the wet state, bridge across the two fractured faces, Fig. 10. Fibre fracture does not seem to have occurred here. This obviously has imparted considerable more toughness to the composites in the wet condition than in the dry condition.

Finally, it is appreciated that in order to obtain definite values for the contribution of the fibre

to the flexural strength and fracture toughness of the composite, tests must be performed on individual fibres in both dry and wet states. Until such results become available it is not possible to attempt a theoretical analysis of the experimental strength and toughness results. However, it will be correct to say that the wet samples mainly derive their toughness from fibre deformation and fibre pull-out. Work absorption in fibre fracture is insignificant. On the contrary, the sources of toughness for the dry samples come from fibre fracture work and fibre pull-out work of both broken and unbroken fibres. These two quantities may be comparable in magnitude. In our earlier work [1] for dry cellulose-cements we suggested that fibre pull-out work is the major source of toughness. This will have to be modified in the light of the experimental results reported here.

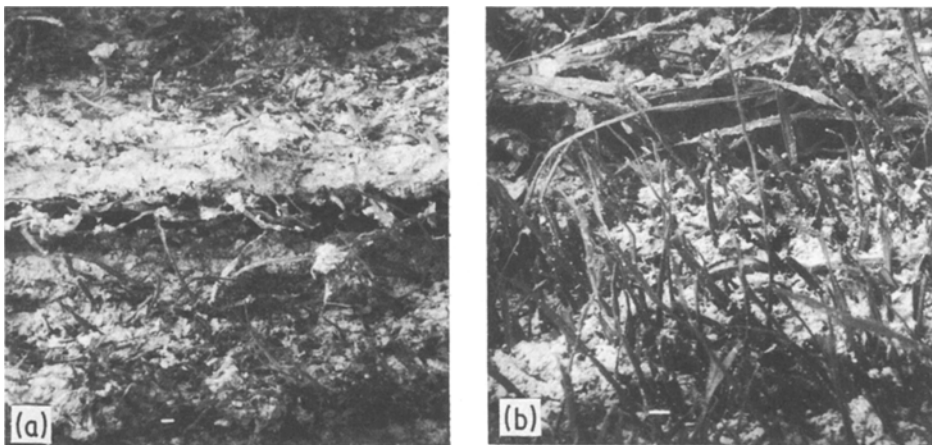


Figure 9 Fracture surfaces of dry Kinleith cellulose-cement composite tested in the flat-wise direction. (a) Top view (25 ×) and (b) 45° tilt (40 ×).

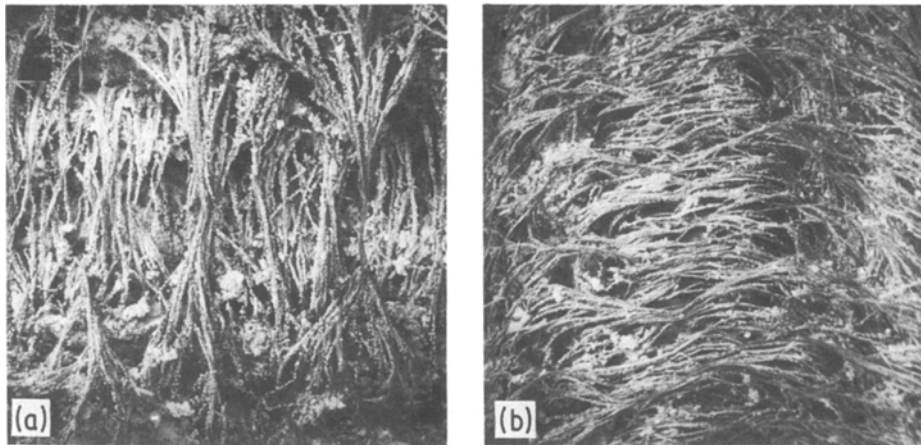


Figure 10 Fracture surface of Kinleith cellulose-cement composite tested in flat-wise direction under wet conditions. (a) Top view (18 X) and (b) 45° tilt (12 X).

## 5. Conclusions

The mechanical behaviour of bleached and unbleached cellulose fibre cements have been compared. It is evident that bleached fibres produce better strength boards in both dry and wet conditions. However the fracture toughnesses are lower than those of unbleached cellulose fibre cements. Water has a dominant effect on the mechanical properties of both types of cellulose fibre cement boards, i.e. Wisakraft and Kinleith kraft fibres. Its presence causes a reduction in strength but an increase in toughnesses. SEM study on the control Kinleith fibre composite has indicated that this effect is the result of the physical properties of the fibre in the two states. In dry conditions, cellulose fibres show a brittle-like behaviour in these composites, in contrast to the wet state, where the toughness of the composite increases due to the increased ductility of the cellulose fibres.

## Acknowledgements

We wish to thank James Hardie & Coy Pty. Ltd

for financially supporting this work and Messrs J. Pether and J. Simpson for useful discussions on the project.

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Received 12 October

and accepted 13 December 1982