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Effect of fibre curl on the properties of wood pulp fibre–cement and silica sheets

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Curl has been induced in unbleached softwood kraft pulp fibres by treatment in the laboratory at 20% consistency in a planetary mixer. Steam treatment of the fibres to set the curl more strongly was found to be detrimental to fibre properties as deduced from handsheet properties. A means of producing two-ply specimens in the laboratory was devised and a tensile test for interlaminar bond strength was developed. The use of curly fibres in reinforced cement and silica sheets gave sheets with improved wet interlaminar bond strengths, relative to sheets prepared from conventionally treated fibres but had little effect on the values of modulus of rupture and fracture toughness.

1. Introduction

Wood pulp fibres are used in a wide range of composite materials [1] and have displaced asbestos fibres in commercial cement sheet products in Australia [2]. These products are composed of multiple thin plies of dewatered fibre–cement–silica slurry subsequently cured in a steam autoclave [3]. They have acceptable properties but the levels of certain properties, particularly of wet sheets, could be usefully improved.

The forces bonding wood pulp fibres to cement matrices are not known with certainty. It has been proposed, however, that hydrogen bonding plays an important role [4]. Such bonds would be expected to be destroyed when exposed to high levels of moisture. A lowering of the modulus of rupture of wood pulp fibre–cement sheets on saturation with water is observed and also a changed mode of fracture from one in which fracture of the fibres predominates to one dominated by their pulling out [5]. Both these changes are consistent with a lowering of the bonding by the action of water. The presence of some broken fibres in the fracture surfaces of wet sheets [5] indicates that there must be maintenance of some bonding of the fibres to the cement either by way of water resistant bonds or by mechanical anchoring of the fibres during pull out [6]. Attempts to increase the wet strength of fibre cement sheets by chemical pretreatment of the wood pulp fibres have proved unsuccessful [7]. The wet strength might, however, be improved if a means could be found of increasing the mechanical anchoring of the fibres.

A similar problem of lack of mechanical strength has been encountered in lightweight paper webs during the early stages of water removal where it has been observed that webs composed of curly fibres have greater resistance to rupture than those composed of straight fibres [8]. If curly fibres were used to reinforce cement sheets then mechanical anchoring of the fibres to the cement might supplement the hydrogen bond forces in the dry sheets and remain in the wet sheets. Curly fibres might also lead to improve interlaminar strength in multi-ply sheets since this property might be expected to be sensitive to the extent of intermingling of the fibres across the interface.

Curl is introduced when the fibres are subjected to strong shear forces while the pulp is at a high consistency [9, 10]. It has been suggested [11] that the fibres form nodules and that curl is a response to shear stresses in the nodules. Whether the introduced curl is permanent or is lost when the pulp suspension is diluted to a low consistency and further processed depends on the type of pulp and the nature of the processing [12]. Mechanical pulp fibres remain curly when disintegrated in cold water but become straight when disintegrated in hot water [13]. The fibres of high yield chemical pulps become straight after only a cold disintegration [11] whereas curl in a low yield chemical pulp fibre appears to be better retained [12]. Recently it has been shown that curl can be set in high yield pulp fibres by a simple steaming treatment [8].

The aims of this work were to examine means of inducing curl in low yield *Pinus radiata* kraft pulp fibres of the type used in cement sheet products [4], the need for setting this curl, and its effect on product properties. This required the development of a method for making a two-ply fibre cement specimen and a test for measuring the interlaminar bond strength. Other properties such as modulus of rupture and fracture energy have also been measured.

2. Experimental procedure

2.1. Induction of curl

The wood pulp fibres used in the experiments were from *Pinus radiata* kraft pulp (Kappa No. approx. 22) ex NZ Pulp and Paper Ltd, Kinleith, New Zealand. 360 g o.d. pulp was soaked in water overnight, given 20 minutes circulation in a Valley beater then beaten for the required time. For each experiment, a con-

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trol sample was retained for sheet-making while the remainder was dewatered by filtering and pressing to approximately 20% consistency. The pulp was then curled for 90 min in a Crypto Peerless E20 mixer, sealed in a plastic bag and placed in a refrigerator. Samples of pulp for the curl-setting experiments were removed from the bags and placed in a steam autoclave at 150° C for 10, 30 or 60 min.

2.2. Handsheet preparation and testing

Handsheets were prepared according to Australian Standard Method AS1301.203S-80.

Zero span tensile index was measured on rewetted samples by using a Pulmac Zero Span Tester according to the maker's handbook. Tensile index and stretch were measured on an Instron 1026 testing machine (Load cell 500 N, Crosshead speed 5 mm min^{-1}) by using samples 15 mm wide and 100 mm in length between TAPPI clamps. Tear index was measured according to Australian Standard Method AS1301.400m-73.

2.3. Fibre–cement–silica composites–preparation and testing

Fibres (8%m/m, on solids), were stirred in water while a cement–silica mixture (1 : 1) was added and the mix was then dewatered and sheets prepared by published methods [14]. Specimens were cut from the sheets and either equilibrated at 22 ± 2° C and 50 ± 5% RH, or saturated with water, and tested in flexure as described previously [14]. The values of fracture toughness were obtained from the areas under the load–deflection curves.

2.4. Two-ply composites

The slurry for making laboratory two-ply composites was divided into two batches each slurried in 300 mL of water. Each sample was stirred for 5 min and poured into a 125 × 125 mm^2 evacuable casting box so that it could be distributed over a screen. The box was evacuated until the sheet appeared dry on the surface and was then tamped carefully. A further vacuum (60 kPa, gauge) was applied for 2 min. The sheet was then removed on the filter screen. Two sheets were made in this manner with the first being stored temporarily with its screen between steel plates in a sealed plastic bag. The screen was removed from the face of the second sheet and the top side of the first sheet was placed against it as shown below in Fig. 1.

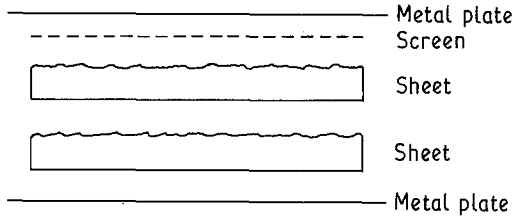

Figure 1 Schematic of formation of two-ply composite.

A stack of eight two-ply sheets was thus assembled and then pressed for 5 min at a pressure of 3.2 MPa. After pressing the screens were removed carefully and the sheets stacked flat in sealed plastic bags for 24 h. Sheets were cured in an autoclave for 8 h at 0.86 MPa steam pressure.

2.5. Interlaminar bond test

A jig was made to fit the 50 kN tension-compression cell on an Instron Model 1114 testing machine and provide areas of 44 × 44 mm^2 for the test. Specimens were cut to size and given a light sand blasting to remove loosely adhering surface particles and a thickness of approximately 1 mm of a two-pot mix of Araldite K125 gap-filling adhesive paste was used to bond the fibre cement sheets to the metal supports, the faces of which had been mechanically roughened. The assemblies were placed in an oven at 100° C for 16 h, allowed to equilibrate at 22° C and 50% RH, and then tested as shown in Fig. 2. For each of the tests described above two specimens were taken from four sheets.

3. Results and discussion

3.1. Fibre curl and handsheet properties

It has been shown [15] that curl can be introduced conveniently into papermaking fibres in the laboratory by treatment at 20% consistency in a Hobart-type kitchen mixer and this was the procedure adopted here. A time of 90 min was chosen because other workers [8] had shown that this was sufficient treatment to produce substantial curliness in a softwood unbleached low-yield kraft pulp. Photomicrographs of the curly fibres and ordinary *P. radiata* unbleached kraft pulp fibres are given in Fig. 3.

Curl indices are best measured by using image analysis [16]. However, indications of the successful introduction of curl into fibres can also be had from changes in the physical properties of handsheets [17].

The effects on the curly fibres of steam treatment at 150° C were also investigated. This treatment had been suggested previously [8] for setting curl in high yield pulp fibres.

The properties of handsheets made from the fibres subjected to the curlation and steam treatments are shown in Table I.

The zero span tensile index was measured on rewetted samples in which the interfibre bonds are largely destroyed and thus is a measure of the fibre rather than the sheet property. The tensile index is a measure of the latter.

The only small variations in zero span tensile index of the pulps, not subjected to steaming, indicates that the heating and curlation treatments had little effect on fibre strength. The higher values of stretch at lower

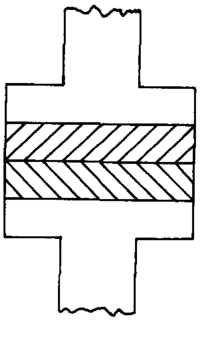

Figure 2 Interlaminar bond test.

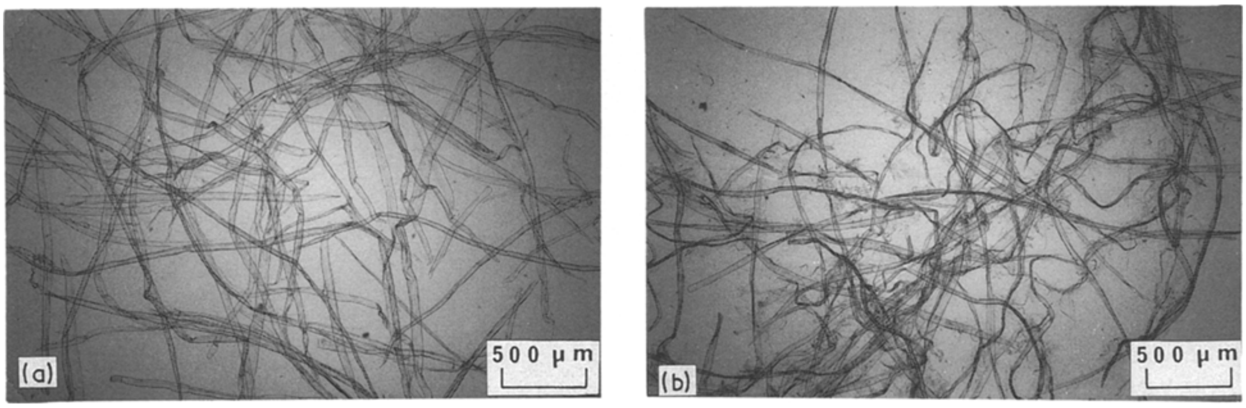

Figure 3 Photomicrographs of (a) unbeaten fibres and (b) curly fibres.

tensile indices of the curly fibres indicate increasing fibre curl rather than improved bonding since values of fibre stretch and tensile index normally increase together with better bonding. The tear index is also increased with the curlier fibres allowing the paper to stretch more before breaking. The markedly reduced values of the tensile index probably arise from the effect of curl which reduces the effective length of the fibres along which stress is transmitted giving, according to the Page equation, a lower tensile strength [18].

The loss in zero span tensile index and greater loss of the tensile index of handsheets made from the steamed fibres might indicate some loss of fibre strength through acid hydrolysis but the increased relative magnitude of the loss of tensile index suggests a loss of interfibre bonding capability as well. There may also be an effect resulting from a change to a more three-dimensional array of fibres as indicated by freer draining pulps.

A further experiment was undertaken to determine whether the observed effects of steam treatment were related to the possible drying out of the pulp in the autoclave. The results of this experiment based on unbeaten pulp are given in Table II. The results show that the degradation of the properties of handsheets made from the steamed pulps was little affected by the addition of excess water.

In addition to possible fibre degradation the increase in freeness caused by the steaming treatments was also undesirable and would make the treated pulps almost unusable on a Hatschek machine [19].

While this work was in progress Page *et al.* [17] showed that the mean curl of a sample of softwood unbleached kraft fibres treated at 20% consistency in a Hobart-type kitchen mixer attained a value typical of curly fibres after 90 min and that the curl was well retained without the use of a setting treatment. This level of treatment was, therefore, chosen for the fibres to be added to the cement and they were used without a setting treatment. Handsheets made from the treated fibres showed increases of 44% in tear index and 28% in stretch and reductions of 30% in tensile index relative to handsheets made from untreated fibres.

3.2. Properties of commercial fibre–cement and silica sheets

The mechanical properties of some commercial fibre-cement sheets made on Hatschek machines are shown in Table III. The asbestos–cellulose (8%/7%, m/m) sheets, now superseded in Australia, are denser and absorb less water on soaking. The interlaminar bond strength of a cellulose-containing dry sheet is about one half that of an asbestos–cellulose reinforced sheet, however, the interlaminar bond strength of a water saturated cellulose fibre sheet is very much less than that of the asbestos containing sheet. Furthermore, the loss of interlaminar bond strength (on saturating the cellulose fibre sheet) is much greater than the concomitant loss of modulus of rupture (MOR) which drops typically to about 65% of its dry value. The magnitude of the loss observed on wetting is reminiscent of the strength loss observed in paper on wetting. In paper, this loss is caused by the breaking of hydrogen bonds by water. This result indicates that the interlaminar bonds are, largely, of the hydrogen bond type.

TABLE I Effect of beating, curl treatment for 90 min and steaming (150° C) on handsheet properties

Pulp treatment	Freeness (mL, CSF)	Zero span tensile index ($N m g^{-1}$)	Tensile index ($N m g^{-1}$)	Stretch (%)	Tear index ($mN m^2 g^{-1}$)
Beaten,	415	178	72.6	3.2	18.4
Unbeaten + curled	565	157	39.5	3.8	25.1
Beaten + curled	440	167	50.1	4.1	26.5
Beaten + curled + steamed (10 min)	620	127	24.3	2.7	22.2
Beaten + curled + steamed (30 min)	600	133	22.5	2.8	16.5
Beaten + curled + steamed (60 min)	630	123	23.4	2.6	16.9

TABLE II Effect of increasing the level of moisture on the properties of handsheets made from curled (90 min) and steamed (15 min) fibres

Pulp treatment	Freeness (mL, CSF)	Zero span tensile index ($\mathrm{N\,m\,g^{-1}}$)	Tensile index ($\mathrm{N\,m\,g^{-1}}$)	Stretch (%)	Tear index ($\mathrm{mN\,m^2\,g^{-1}}$)
Unbeaten and curled	565	157	39.5	3.8	25.1
Curled + steamed	675	122	19.7	2.2	17.4
Curled + water added + steamed	665	127	21.0	2.0	17.3

The actual values shown in Table I may be an underestimate of the true values to the extent that the values are reduced by edge flaws introduced in cutting $44 \times 44\,\mathrm{mm}^2$ samples. Thus larger samples might give somewhat higher values though there is no reason to believe that the relative values would be changed to an appreciable extent.

The values of modulus of rupture and of fracture toughness of the cellulose fibre reinforced sheets are only marginally below the corresponding values for the mixed asbestos–cellulose fibre reinforced sheets.

Scanning electron micrographs of the interlaminar failure surfaces of wet and dry samples of both types of sheets are shown in Fig. 4.

The differences in fineness between the asbestos and cellulose fibres are apparent. Little can be said from these micrographs, about differences in the original planarity of the fibres because the fibres will have been raised from the surface during straining in the test.

3.3. Comparison of properties of one- and two-ply sheets

In Table IV the properties of the two-ply sheets made by the new technique are compared with those of the one-ply sheets made by the standard technique. The new sheets are slightly less dense and absorb slightly more water, have similar modulus of rupture values for dry and water-saturated samples and slightly higher fracture toughnesses. The interlaminar bond strengths of the two-ply sheets in both the dry and wet conditions are lower than the internal bond strengths of the one-ply sheets but above the values for the commercial multiply sheets. The laboratory and commercial sheets are produced under different conditions and have different fibre orientations. Moreover, the multi-ply nature of the commercial sheets must increase the probability of the presence of weak interply bonds.

3.4. Properties of curly fibre–cement and silica sheets

The mechanical properties of sheets comprising cement and essentially unbeaten fibres, fibres beaten to 250 mL CSF in a Valley beater, unbeaten fibre curlated as described earlier and fibres curled following beating to 250 mL CSF are shown in Table V. The average densities of the sheets are in the range 1.30 to 1.35 and the average water absorptions in the range 32.8 to 34.9. The values of modulus of rupture (MOR) and fracture toughness of the sheets appear to be not greatly affected by beating or curling the fibres, however, the wet interlaminar bond strengths do appear to be improved significantly when curly fibres are used and are of the order of the internal bond strengths of one-ply sheets (see Table IV). Whether this result would be fully reproduced on sheets made on a Hatschek machine is problematical since the untreated laboratory sheets did not show the same loss of bond strength on soaking as did the machine-made sheets.

Microscopic confirmation of the retention of curl by the fibres in the sheets has proved difficult because of the covering matrix material. Attempts to remove this material by etching with strong acid [20] have not been successful. The only fibres lightly covered by matrix are on the edges. Fig. 5 shows fibres in one such edge forming a nodular structure reminiscent of those found in curly fibres (Fig. 3). The micrographs do not

TABLE III Properties of commercial fibre–cement–silica sheets

Sheet	Condition	Density ($\mathrm{g\,cm^{-3}}$)	Water absorbing (%)	Interlaminar bond strength (MPa)	MOR[a] (MPa)		Fracture toughness[a] ($\mathrm{J\,m^{-2}}$)	
Asbestos–cellulose	50% RH	1.41 ± 0.01	26.4 ± 0.5	1.62 ± 0.2	27.3 ± 0.85 15.2 ± 1.0	20.3[b] ± 0.8	1785 ± 85 1030 ± 20	1355[b] ± 45
Asbestos–cellulose	Sat[c]			0.82 ± 0.1	17.3 ± 0.6 11.0 ± 0.4	13.4[b] ± 0.5	3340 ± 80 1160 ± 80	1970[b] ± 90
Cellulose	50% RH	1.30 ± 0.01	34.0 ± 0.5	0.85 ± 0.1	23.8 ± 1.0 15.5 ± 0.3	19.2[b] ± 0.6	1645 ± 85 945 ± 40	1245[b] ± 60
Cellulose	Sat[c]			< 0.10	15.9 ± 0.4 9.6 ± 0.2	12.4[b] ± 0.3	2740 ± 90 1060 ± 35	1705[b] ± 55

[a] Upper value is for machine direction, lower value is for cross direction.

[b] Geometric mean of values for machine and cross directions.

[c] Saturated with water.

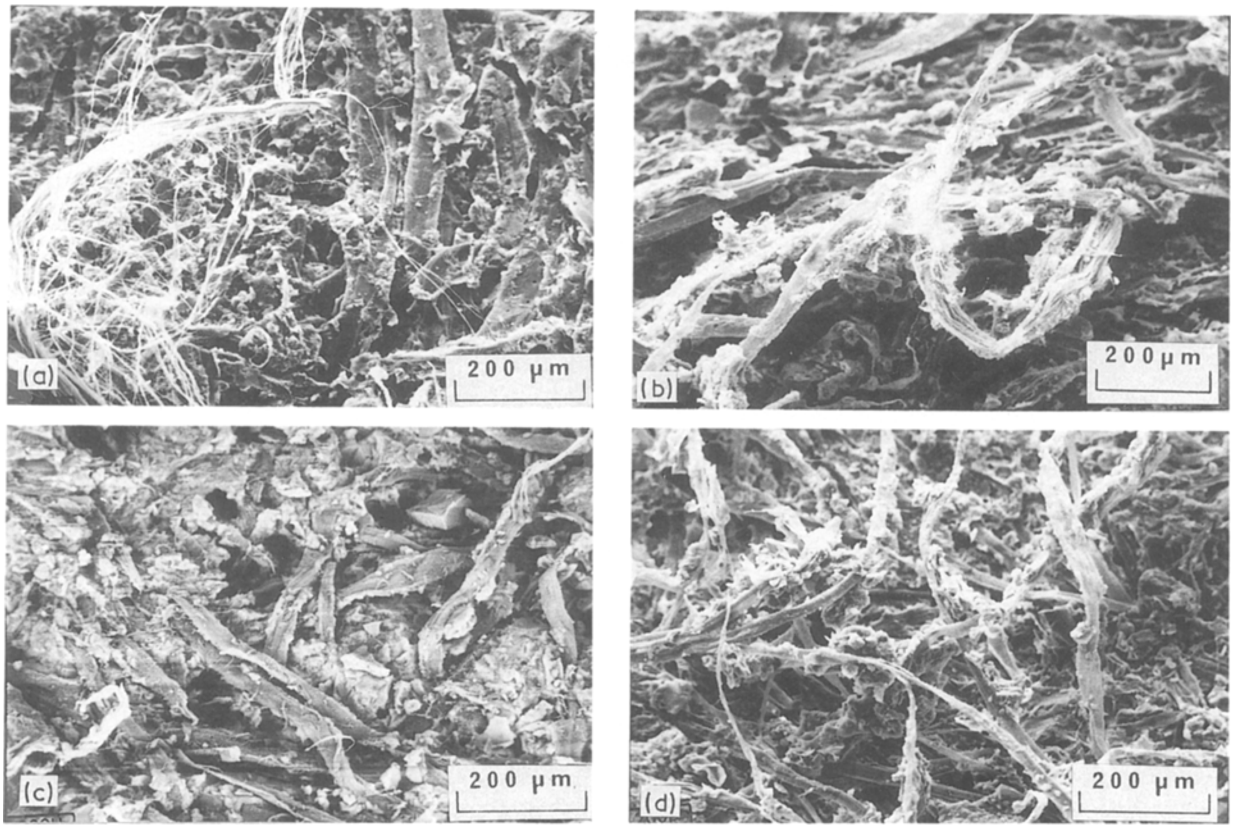

Figure 4 SEMs showing tensile interlaminar fracture surfaces of commercial fibre–cement–silica sheets. (a) Dry asbestos–cellulose fibre sheet. (b) Wet asbestos–cellulose fibre sheet. (c) Dry cellulose fibre sheet. (d) Wet cellulose fibre sheet.

enable us to distinguish between two possible changes brought about by having curly fibres, namely, their lying out-of-plane and bridging the laminae and the effect of their greater three dimensionality in reducing the possibility of a fibre-rich cement-lean layer occurring at the interface.

4. Conclusions

Curl can be induced in unbleached *P. radiata* kraft pulp fibres by mechanical treatment at 20% consistency. Attempts to set this curl more strongly by steam treatments had a detrimental effect on the mechanical properties of the fibres as deduced from the properties

TABLE IV Properties of laboratory kraft pulp unbeaten fibre–cement–silica one- and two-ply sheets

Sheet	Condition	Density (g cm^{-3})	Water absorbing (%)	Internal or interlaminar bond strength (MPa)	MOR (MPa)	Fracture toughness (J m^{-2})
One-ply	50% RH	1.38 ± 0.03	31.3 ± 1.2	3.1[b] ± 0.2	23.2 ± 2.1	1285 ± 110
	Sat[a]			1.4[b] ± 0.2	15.1 ± 1.5	2750 ± 190
Two-ply	50% RH	1.34 ± 0.02	32.8 ± 0.8	2.4[c] ± 0.2	24.6 ± 1.3	1785 ± 125
	Sat[a]			1.0[c] ± 0.2	15.1 ± 1.0	2840 ± 115

[a] Saturated with water.
[b] Internal bond strength.
[c] Interlaminar bond strength.

TABLE V Properties of curled kraft pulp fibre–cement–silica two-ply sheets

Sheet	Condition	Density (g cm^{-3})	Water absorbing (%)	Interlaminar bond strength (MPa)	MOR (MPa)	Fracture toughness (J m^{-2})
Unbeaten	50% RH	1.34 ± 0.02	32.8 ± 0.8	2.4 ± 0.2	24.6 ± 1.3	1785 ± 125
	Sat[a]			1.0 ± 0.2	15.1 ± 1.0	2840 ± 115
Beaten	50% RH	1.35 ± 0.03	34.0 ± 1.9	2.6 ± 0.2	22.0 ± 1.2	1550 ± 150
	Sat[a]			0.9 ± 0.2	16.1 ± 1.2	3060 ± 360
Curlated	50% RH	1.30 ± 0.04	34.9 ± 2.3	3.0 ± 0.3	22.1 ± 1.2	1525 ± 65
	Sat[a]			1.5 ± 0.2	13.8 ± 0.7	2620 ± 235
Beaten and curlated	50% RH	1.35 ± 0.03	33.5 ± 1.8	2.6 ± 0.4	21.6 ± 1.6	1895 ± 240
	Sat[a]			1.8 ± 0.4	15.6 ± 1.5	3025 ± 590

[a] Saturated with water.

Figure 5 SEM of edge of fibre–cement–silica sheet containing curled fibres.

of handsheets containing them and made them much freer draining. Fibres curled in this way when used to reinforce cement–silica sheets give sheets showing improvement in wet interlaminar tensile strength and with values of modulus of rupture and fracture toughness similar to those of beaten fibres.

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References

1. P. ZADORECKI and A. J. MICHELL, *Polym. Compos.* **10** (1989) 69.
2. CSIRO Industrial Research News. No. 146 (May 1981).
3. R. S. P. COUTTS and A. J. MICHELL, *J. Appl. Polym. Sci.: Polym. Symp.* **37** (1983) 829.
4. R. S. P. COUTTS and V. RIDIKAS, *Appita* **35** (1982) 395.
5. R. S. P. COUTTS and P. KIGHTLY, *J. Mater. Sci.* **17** (1982) 1801.
6. F. E. MORRISSEY, R. S. P. COUTTS and P. U. A. GROSSMAN, *Int. J. Cem. Compos. Lightweight Concr.* **7** (1985) 73.
7. R. S. P. COUTTS, unpublished results.
8. M. C. BARBE, R. S. SETH and D. H. PAGE, *Pulp Pap. Can.* **85** (1984) 65.
9. J. H. De GRACE and D. H. PAGE, *Tappi* **59** (7) (1976) 98.
10. D. H. PAGE and R. S. SETH, *Pulp Pap. Can.* **80** (1979) T235.
11. H. S. HILL, J. EDWARDS and L. R. BEATH, *Tappi* **33** (1950) 36.
12. D. H. PAGE, M. C. BARBE, R. S. SETH and B. D. JORDAN, *J. Pulp Pap. Sci.* **10** (1984) 74.
13. H. W. H. JONES, *Pulp Pap. Mag. Can.* **67** (1966) T283.
14. R. S. P. COUTTS, *Composites* **15** (1983) 139.
15. D. H. PAGE, R. S. SETH and J. H. De GRACE, *Tappi* **62** (1979) 99.
16. B. D. JORDAN and D. H. PAGE, in "Role of Fundamental Research in Papermaking", Cambridge September 1981, edited by J. Brander (Mechanical Engineering Publications Ltd., London, 1983) p. 745.
17. D. H. PAGE, R. S. SETH, B. D. JORDAN and M. C. BARBE, in "Papermaking Raw Materials", Oxford, September 1985, edited by V. Punton (Mechanical Engineering Publications, London, 1985) p. 183.
18. D. H. PAGE, *Tappi* **52** (1969) 674.
19. A. M. COOKE, personal communication.
20. Y. W. MAI, M. I. HAKEEM and B. COTTERELL, *J. Mater. Sci.* **18** (1983) 2156.

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